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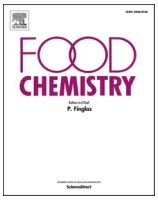

NMR-based approach to detect white wine vinegar fraud

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ABSTRACT

Low-Field Nuclear Magnetic Resonance (LF-NMR) can be a valid tool in food fingerprint analyses to detect commercial frauds. Thus, the work aims at exploring the potential of LF-NMR, coupled with chemometrics, in discriminating authentic white wine vinegars from products adulterated with alcohol vinegars (i.e., 5–25% *v*/v adulteration levels). The monodimensional spectra and transverse relaxation times (T2) of 88 samples, including 32 authentic vinegars and 56 adulterated samples, were collected. Three different spectral regions were investigated (i.e., 3.75–0.90, 3.75–2.00, and 1.50–0.90 ppm) and, for each, fifteen variables were selected from the pretreated monodimensional spectra. Linear Discriminant Analysis (LDA) on monodimensional spectra in the range 3.75–0.90 ppm gave 100% correct classification of authentic and adulterated vinegars in prediction, whereas LDA models developed with acetic acid or water T2 failed.

In conclusion, LF-NMR spectra can be effectively used to detect, in a rapid and non-destructive way, white wine vinegar adulteration with alcohol vinegar.

1. Introduction

Within the food industry, vinegar is a product particularly prone to fraudulent manipulation. Several types of vinegars are produced worldwide with distinctive characteristics based on raw materials and production methods. A substantial share of the worldwide vinegar market is covered by Balsamic vinegar, red wine vinegar, and white wine vinegar (Bekatorou, 2019). In the Mediterranean Countries and Central Europe, which are major wine manufacturing regions, wine vinegar is the most commonly used vinegar.

Economic frauds are particularly common in the field, from the substitution of aged balsamic vinegars with its cheaper alternatives to the dilution of vinegar with diluted synthetic acid (Grégrová, Čížková, Mazáč, & Voldřich, 2012). The addition of low-value products, for example spirit vinegar, to products of higher economic value, such as wine vinegar, turns out to be one of the most frequently common frauds (Callejón et al., 2018). Indeed, spirit vinegars are characterized by similar macronutrient composition, but they are made by fermentation of alcohol derived from starchy or saccharides-containing raw materials (e.g., molasses, potatoes or grain alcohol) (Grégrová et al., 2012), even though information about raw materials are not mandatory in the labels. Frauds are particularly relevant to the canning industry where the volumes of vinegar used are large and this fraudulent practice can have a substantial economic impact, causing damages to both buyers and honest manufacturers. With respect to this issue, the food industry requires solid and reliable methods to authenticate vinegar, which are also quick and inexpensive. Often the diffusion of a new methodology is hampered by the complex sample preparation and the associated costs. Therefore, methods that do not require any (or minimal) sample treatment are highly desired. However, the increasing number of adulterants used in frauds makes the process of authentication extremely complex.

In this context, Nuclear Magnetic Resonance (NMR) spectroscopy turns out to be one of the most interesting analytical tools: in addition to the possibility of making quantitative determinations, it is possible to simultaneously detect several chemical compounds within the analyzed matrix through non-destructive, rapid analyses that often do not require any sample preparations (Consonni & Cagliani, 2019; Pacholczyk-Sienicka, Ciepielowski, & Albrecht, 2021). Depending on the needs of the investigation being conducted, NMR analyses can be carried out by targeted and non-targeted approaches. In general, both the approaches proceed according to metabolomics. The targeted approach is useful when the determination of specific markers, known beforehand, is needed. However, it is not always possible to define a priori markers of food adulteration. Thus, it is often useful to resort to non-targeted methods: the focus here falls on pattern recognition, which can distinguish samples according to their class of membership, based on the

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simultaneous analysis of several compounds or the metabolic profile of the analyzed matrix (Medina, Pereira, Silva, Perestrelo, & Câmara, 2019; Mialon, Roig, Capodanno, & Cadiere, 2023). In the field of food authentication, given the potential of NMR analysis to generate a large amount of data in a relatively short period of time, the use of the untargeted approach in combination with chemometric techniques has great advantages, and consequently there are numerous studies in this regard on various food matrices, such as meat (Jakes et al., 2015; Leng et al., 2023), honey, beer, spices (Kuballa, Brunner, Thongpanchang, Walch, & Lachenmeier, 2018), fruit juice (Cuny et al., 2008), wine (Amargianitaki & Spyros, 2017), and vinegar (Consonni et al., 2008; Hsieh, Li, Cheng, & Ma, 2013; Mascareli et al., 2023). Most studies, however, focus on the use of NMR techniques by high-field (HF-)instruments, while the low-field (LF-)NMR instruments, i.e. time-domain NMR (TD-NMR), have several advantages. The functioning of HF-instruments relies on superconducting magnets that must necessarily be cooled with cryogenic liquids, significantly affecting the operational costs of analysis. LF-instruments, on the other hand, use permanent magnets that, in addition to having lower operating costs, allow these instruments to be significantly smaller than HF-instruments and therefore usable as bench equipment in virtually any laboratory. LF-instruments are also definitely cheaper with respect to HF-equipment, being their cost about eight times lower (Pagès et al., 2014). However, the lower field strength of LF-instruments results in lower sensitivity and resolution. Thus, these instruments are best suited for analyses where it is not essential to elucidate the fine structural and chemical details of the compounds under investigation: low-resolution analyses often do not offer chemical shift information, and the ^{1}H signal obtained includes a single absorption that encodes information from all protons in the sample (Baroni, Consonni, Ferrante, & Aime, 2009). However, these disadvantages can be overcome by appropriate coupling of chemometrics techniques (Galvan et al., 2021).

Therefore, this work aims to verify the hypothesis that a LF-NMR instrument, with the support of chemometrics, can be used in rapid untargeted analyses, without any sample preparation, to test for adulteration, at different percentages (5–25% *v*/v adulteration levels), of white wine vinegars with alcohol vinegars. Given the absence of internal references in the NMR spectra, the iCOSHIFT algorithm (Savorani, Tomasi, & Engelsen, 2010) was used for pretreatment, coupled with the Savitzky-Golay algorithm (Savitzky & Golay, 1964) for noise reduction. In addition, with the aim of creating more robust and concise models, ultimately reducing the computational requirements needed, the SELECT algorithm of the V-PARVUS package (Forina et al., 1988) was used for the selection of the most informative variables.

2. Materials and methods

2.1. Authentic and adulterated vinegars

The sample set was created using 32 commercial white wine vinegars (i.e., authentic samples), covering different brands, producers, lots, and origins to represent the product variability in the Italian market, and 2 commercial spirit vinegars (i.e., adulterants).

Details about the vinegars are reported in Table A1. The majority (28%) were produced by the same producer, followed by other seven different companies; the considered samples covered 16 different Italian brands, commercial brands have not been made explicit for the sake of confidentiality. Five main areas of production are indicated in the labels: Modena and Novara (28%), Naples (24% of samples), Mantua (16%), and Arezzo (4%). The two commercial spirit vinegar were from Naples and Arezzo, respectively. Four authentic samples were bought in three different production lots and selected for preparation of adulterated samples. In particular, the three bottles belonging to different lots were used to prepare samples adulterated with 25% *v*/v of each spirit vinegar (for a total of 24 samples adulterated at the highest level), while only one bottle of each selected white wine vinegar was used to prepare samples adulterated with the two spirit vinegars at different levels (i.e., 5, 10, 15, and 20% v/v; for a total of 32 differently adulterated samples). Thus, a total of 88 samples was analyzed in duplicate, including 32 authentic samples and 56 adulterated samples (Fig. 1).

2.2. NMR measurements

The reference spectra of one authentic white wine vinegar were acquired by a HF-NMR instrument (AV600 spectrometer, Bruker Corporation, Billerica, MA, USA) operating at a frequency of 600.10 MHz for ^{1}H, equipped with a z-gradient 5 mm reverse probe. ^{1}H NMR spectra were recorded at 25 °C. Chemical shifts (δ) were measured in ppm and referenced to external 2,2-dimethyl-2-silapentane-5-sulfonate sodium salt set at 0.00 ppm, 0.14% in analyzed samples. The samples were added by 5% of D_2O. Solvent suppression was achieved by presaturation with the carrier placed on the water resonance (4.70 ppm). NMR spectra were elaborated by using the TOPSPIN 1.3 software (Bruker BioSpin GmbH, Rheinstetten, Germany).

The whole set of samples (88 samples) was analyzed in duplicate by a LF-NMR (Spinsolve 60 NMR spectrometer, Magritek Ltd. Aachen, Germany) operating at a resonance frequency of 60.0 MHz. Samples were used directly without any solvent/standard addition. Monodimensional spectra and transverse relaxation times (spin-spin, T2) were recorded. For the spectra acquisition, 32 scans (FIDs) were collected into 8 k data points using 90 pulse angle, acquisition time of 3.7 s, and 10 s repetition time. Phase correction was performed manually for each spectrum, and the baseline correction was applied over the entire spectral range. The monodimensional spectra were processed using MestReNova (v. 12.0.2–20,910, Mestrelab Research S.L, Santiago de Compostela, Spain). The water and acetic acid T2 were acquired applying the Carr-Purcell-Meiboom-Gill sequence (CPMG) (Carr & Purcell, 1954; Meiboom & Gill, 1958). The experimental parameters were as follows: acquisition time 3.2 s, repetition time 15 s, number of steps 20, echo-time 10,000 μs. The exponential decay curve of each relaxation time measurement was the result of the accumulation of 4 scans. An exponential decay curve of the raw data was transformed to a continuous relaxation time distribution curve by inverse Laplace transformation.

2.3. Data analysis

The LF-NMR data collected in duplicate for each sample were averaged and three datasets were created with monodimensional spectra, water T2, and acetic acid T2, respectively.

Prior to data analysis, the spectral dataset was reduced in the region 0.90–3.75 ppm (named region A) according to previous findings (Caligiani, Acquotti, Palla, & Bocchi, 2007; Boffo, Tavares, Ferreira, & Ferreira, 2009). The region A was further divided in two subregions: region B (2.00–3.75 ppm) and C (0.90–1.50 ppm). To correct the inhomogeneous chemical shifts, all spectra were aligned by means of the iCOSHIFT algorithm (Savorani et al., 2010), whereas the Savitzky-Golay algorithm (Savitzky & Golay, 1964) was used for noise reduction (15 points window, 2nd polynomial order).

Principal Component Analysis (PCA) was performed on monodimensional spectra and T2 datasets, enabling the evaluation of variable load and possible sample patterns according to the degree of adulteration.

Subsequently, the datasets were prepared for Linear Discriminant Analysis (LDA) by data split and variable selection. Each dataset was divided in a calibration set (about 70% of the whole collected data) for calibration and cross-validation (with 5 cancellation groups) and an external test set containing only untrained samples (30% of the whole collected data). Data were systematically split into calibration and validation set guaranteeing the same variability of the initial data pool in terms of vinegar producers, origins, adulteration levels, and adulterant. In detail, referring to sample codes reported in Table 1S, the procedure guaranteed the presence in the test set of 9 authentic samples

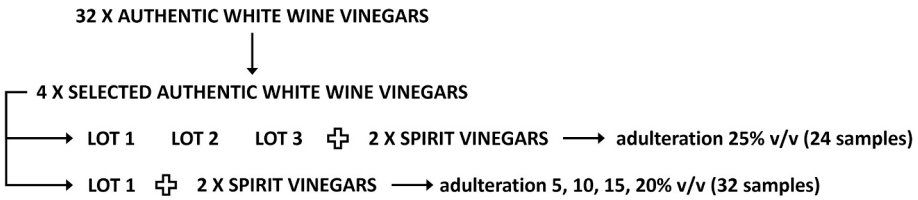

Fig. 1. Schematic representation of the vinegar sample set.

(AC1 x 3 lots, AC6:AC11) and 18 adulterated samples as follows: 9 at 25% adulteration level (3 lot for AC1 with AE1, AC2 with AE2, AC3 with AE1), 2 at 5% (AC2AE1, AC2AE2), 2 at 10% (AC1AE1, AC1AE2), 2 at 15% (AC5AE1, AC5AE2), and 2 at 20% (AC1AE1, AC1AE2).

The LDA modelling was implemented as proper solution for the specific authentication issue since the two considered classes (i.e. authentic and adulterated vinegars) are meaningfully defined and suitably sampled (Oliveri, 2017). As LDA utilizes discriminant canonicals to calculate the centre of matrix covariance, it needs a number of samples exceeding the number of variables. Thus, fifteen variables were selected from the monodimensional spectra dataset by the SELECT algorithm, implemented in V-PARVUS package (Forina et al., 1988). The algorithm selects the variable with the largest Fisher weight, then decorrelates the other predictors; the procedure is repeated iteratively until a fixed number of variables is selected (i.e. 15 variables in this study). The dataset of T2 and the fifteen variables selected from monodimensional spectra were used for LDA, using the V-PARVUS package, to discriminate between authentic and adulterated samples' classes. Model performances were evaluated in terms of correct classification ability in calibration, cross-validation, and prediction.

3. Results and discussion

3.1. Data inspection

A representative ^{1}H NMR profile of wine vinegar is reported in Fig. 2; as expected, the signals associated with the major components (i.e., organic acids, ethanol, sugars, and amino acids) are clearly visible. In particular, it is possible to identify acetic acid (singlet signal at 1.91 ppm), ethanol (triplet signal at 1.16 ppm and quadruplet signal at 4.65 ppm), malic acid (double doublet signal at 2.85 and 2.76 ppm), succinic acid (singlet at 2.45 ppm), and citric acid (two doublet signals at 2.98 and 2.89 ppm), which are the main chemical markers in wine vinegar. Anomeric forms α and β of glucose show signals in the spectral region between 3.0 and 5.5 ppm and are observed in very small intensity. The signals of amino acids are also very small. The assignment of the NMR peaks agrees with previous NMR studies (Caligiani et al., 2007).

In Fig. 3 examples of the LF-NMR monodimensional spectra are reported, divided according to the three regions used in statistical analysis: the aliphatic/alcoholic region A from 3.75 to 0.95 ppm (Fig. 3A); the region B from 3.75 to 2.00 ppm (Fig. 3B); the spectral region C from 1.50 to 0.90 ppm (Fig. 3C). In particular, Fig. 3 shows the LF-NMR spectra of one authentic white wine vinegar and its adulterations from 5 to 25%,

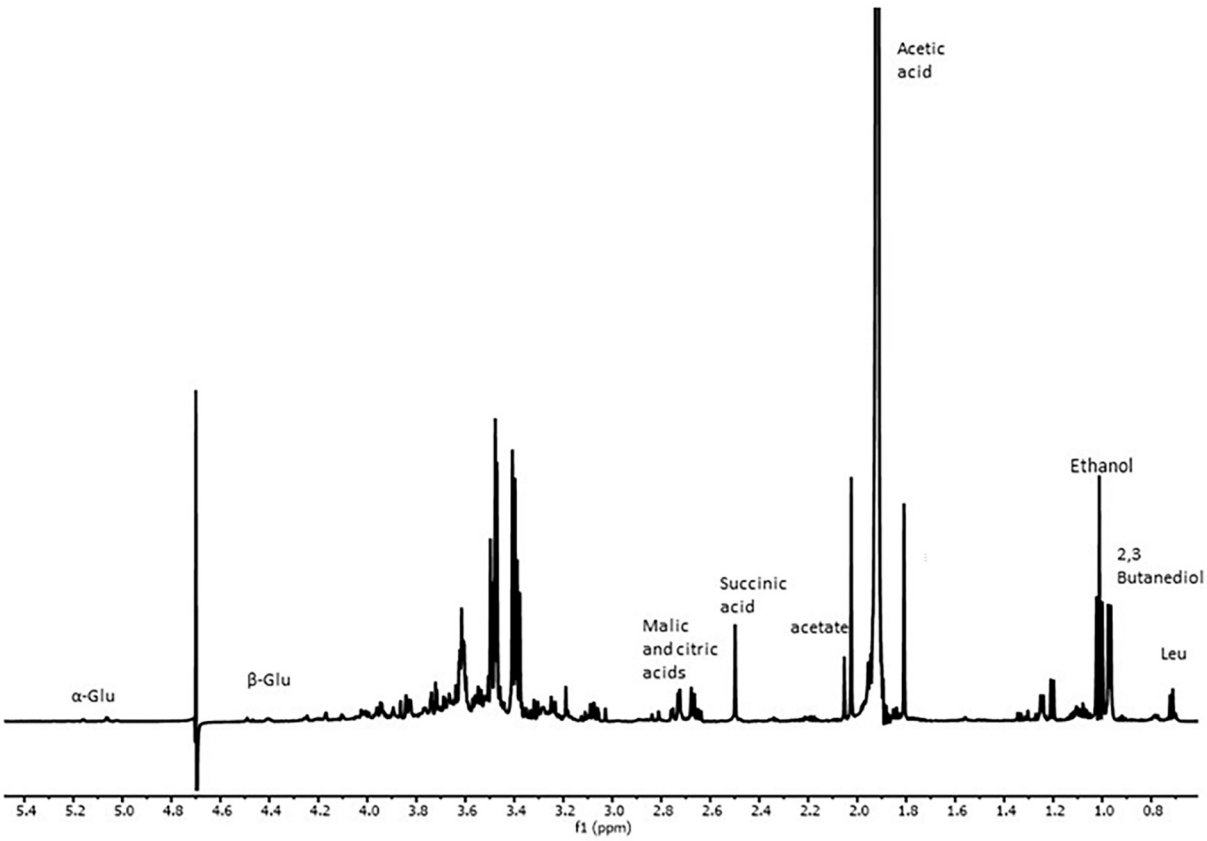

Fig. 2. ^{1}H NMR spectra of authentic white wine vinegar recorded at 600 MHz, with water suppression.

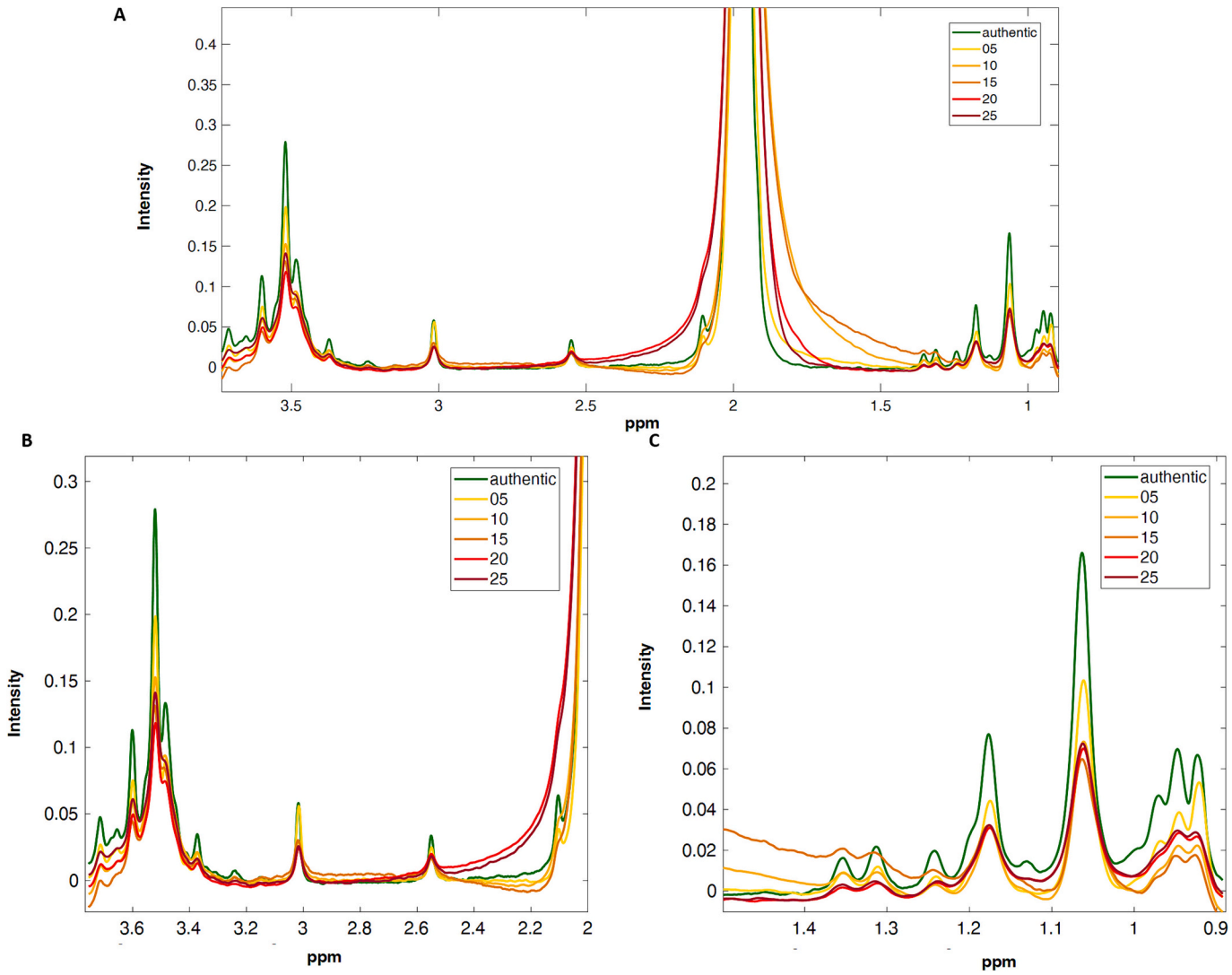

Fig. 3. Selected regions of the LF-NMR monodimensional spectra for one of the authentic white wine vinegars and its adulterations from 5 to 25%. A: aliphatic/alcoholic region from 3.75 to 0.90 ppm; B: region from 3.75 and 2.00 ppm; C: region from 1.50 to 0.90 ppm.

revealing a link between the intensities of certain signals and the degree of adulteration. With the increase in adulteration level, an enlargement of the acetic acid band is observed, leading to the incorporation of the signal at 2.07 ppm, attributable to acetyl-sugar; moreover, all signals generally decrease in intensity. In the region between 3.75 and 3.00 ppm the overlapping signals of the sugar are observed. Well resolved signals are observed at 3.03–2.45 ppm, attributable to citric, malic and succinic acids, respectively. In the adulterated vinegar samples, the LF-NMR profile changes only for the intensity of the signals (Fig. 3B and C), with a dilution and broadening effect as the adulteration levels increase.

Baroni et al. (2009) investigated the interaction of water molecules with vinegar components by measuring the NMR relaxation times, i.e. the spin-lattice (T1) and the spin-spin (T2) relaxation times of water protons; the measurement of T1 and T2 resulted useful for the characterization of aging process and fraud detection. Fig. 4A and C report the trends of T2 for acetic acid and water of one authentic white wine vinegar and its adulterations. To better highlight the differences between authentic and adulterated samples, the T2 values are also shown after natural logarithmic transformation (Figs. 4B and 4D). It is possible to observe that an increase in the adulteration level with spirit vinegar leads to an increase in T2 values and changes in their slope, especially at the highest adulteration level (i.e., 25%).

3.2. Data exploration

The pre-processed dataset of the monodimensional spectra was explored by means of PCA, considering the three different spectral ranges A, B, and C. The PC1 vs PC2 score plot obtained by the PCA performed on the whole aliphatic/alcoholic region A (Fig. 5A) showed a clear separation of the samples adulterated at 25% *v*/v from the other samples. Indeed, samples adulterated at the highest level were characterized by positive PC1 values, whereas all the other samples had negative PC1 scores. The loading plot (Fig. 5B) confirmed what observed in the monodimensional spectra: The main responsible of the sample distribution is the disappearance of the signal attributed to acetyl-sugar (at 2.07 ppm), which is incorporated into the broad peak of acetic acid when the adulteration is at high levels.

The exploratory data analysis on the T2 dataset for acetic acid, after natural logarithm transformation, revealed a good sample distribution according to the adulteration level. Indeed, almost all the authentic samples were characterized by negative PC1 scores (Fig. 5C) and almost all the samples adulterated at 25% v/v had positive PC1 values, whereas the samples adulterated at lower levels (from 5 to 20% *v*/v) are distributed in-between. The loading plot (Fig. 5D) confirmed that the sample pattern is due to the change in slope of the spin-spin relaxation time of acetic acid, as already observed in the data after logarithmic

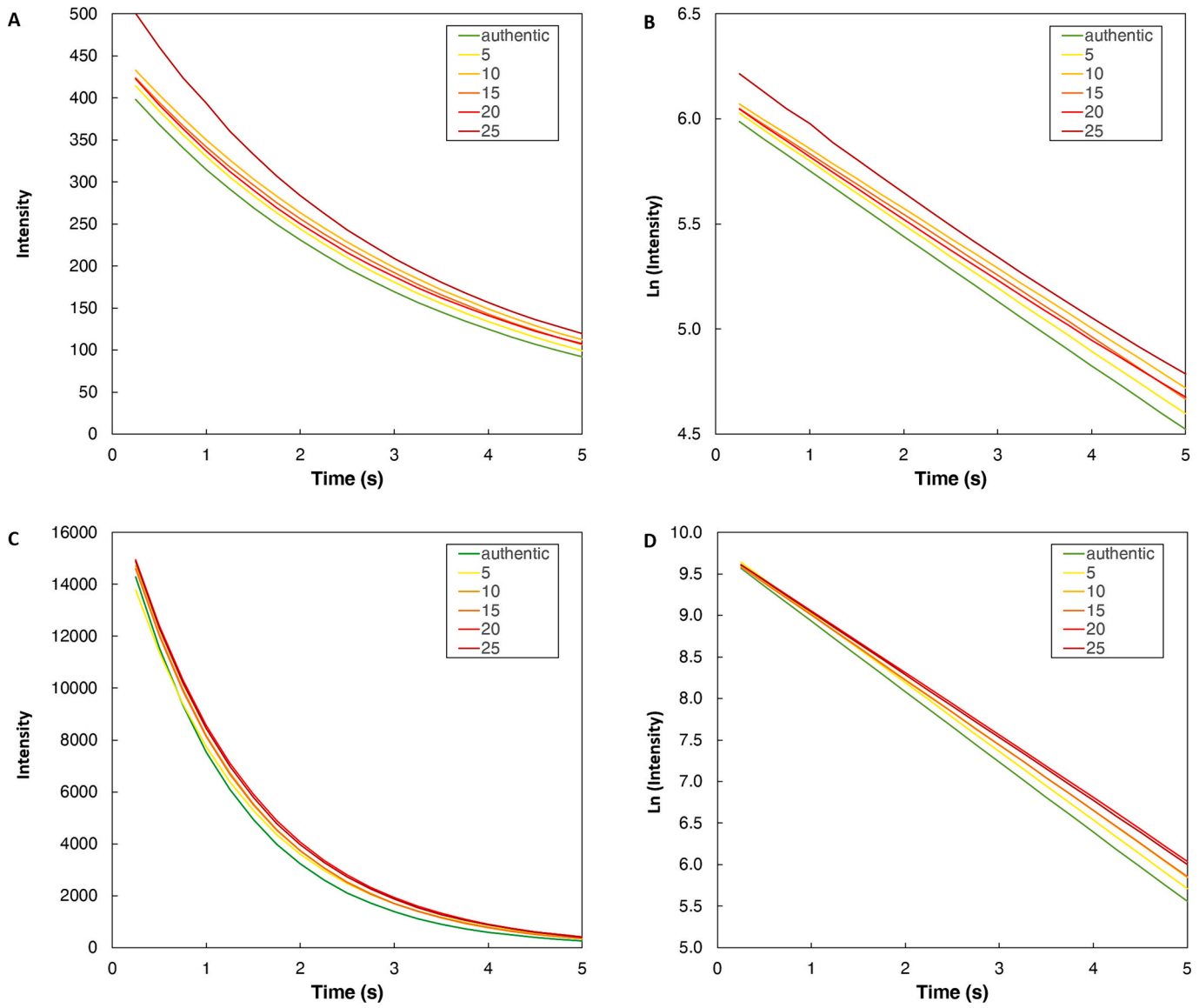

Fig. 4. T2 spin-spin relaxation time for water (A and B) and for acetic acid (C and D) for one of the authentic white wine vinegars and its adulterations from 5 to 25%.

transformation (Fig. 4D). No distribution trends according to adulteration level were observed for T2 of water (data not shown).

From T^2 vs Q residual plot obtained from PCA, few outliers were observed and consequently removed from the datasets used for the construction of the classification models; in details, one outlier was detected for the monodimensional spectra and three for T2.

3.3. Classification models

By using the monodimensional spectral data, the best performance in discriminating authentic and adulterated vinegars was obtained by the LDA model developed with the whole aliphatic/alcoholic region (region A, from 3.75 to 0.95 ppm). All the samples were correctly classified in the a priori assigned class in calibration, cross-validation, and prediction (Table 1). The 15 selected variables were the ones with higher weights in the PCA previously commented (i.e., around 2.00 ppm and 3.5 ppm), attributable to acetyl-sugar, sugar, and glycerol. In detail, the signals around 1.7 ppm have not been discussed in previous literature concerning vinegar. However, they have been previously assigned to citrulline (1.7 and 1.95 δ) in fruit juice analysis (Belton et al., 1996). Furthermore, the variables selected in the range 2.50 to 3.00 ppm could be related to the specific chemical markers of wine vinegar, i.e. malic acid (double doublet signal at 2.85 and 2.76 ppm), succinic acid (singlet at 2.45 ppm), and citric acid (two doublet signals at 2.98 and 2.89 ppm) (Caligiani et al., 2007). Finally, the variables selected from 3.40 ppm to 3.70 ppm are probably related to sugar signals (Caligiani et al., 2007).

The LDA models developed selecting the data of the sub-regions B and C gave a good prediction ability. Indeed, both models gave a 100% correct classification rate in prediction for the authentic samples. However, their cross-validation phases ware not equally performing, thus suggesting less robustness in the model performance for both B and C region.

Not as good results were obtained by modelling T2 data. The models developed with acetic acid or water T2 failed in discriminating authentic from adulterated samples (Table 1). This could be the reason why T2 spin–spin relaxation measurements data have been scarcely used for detection of fraudulent behavior in the vinegar sector (Baroni et al., 2009).

The LDA models developed using the monodimensional spectra regions overcome what have been presented in the scientific literature until now. Indeed, different classification strategies have been applied to model NMR data for fraudulent behavior detection, from agronomic

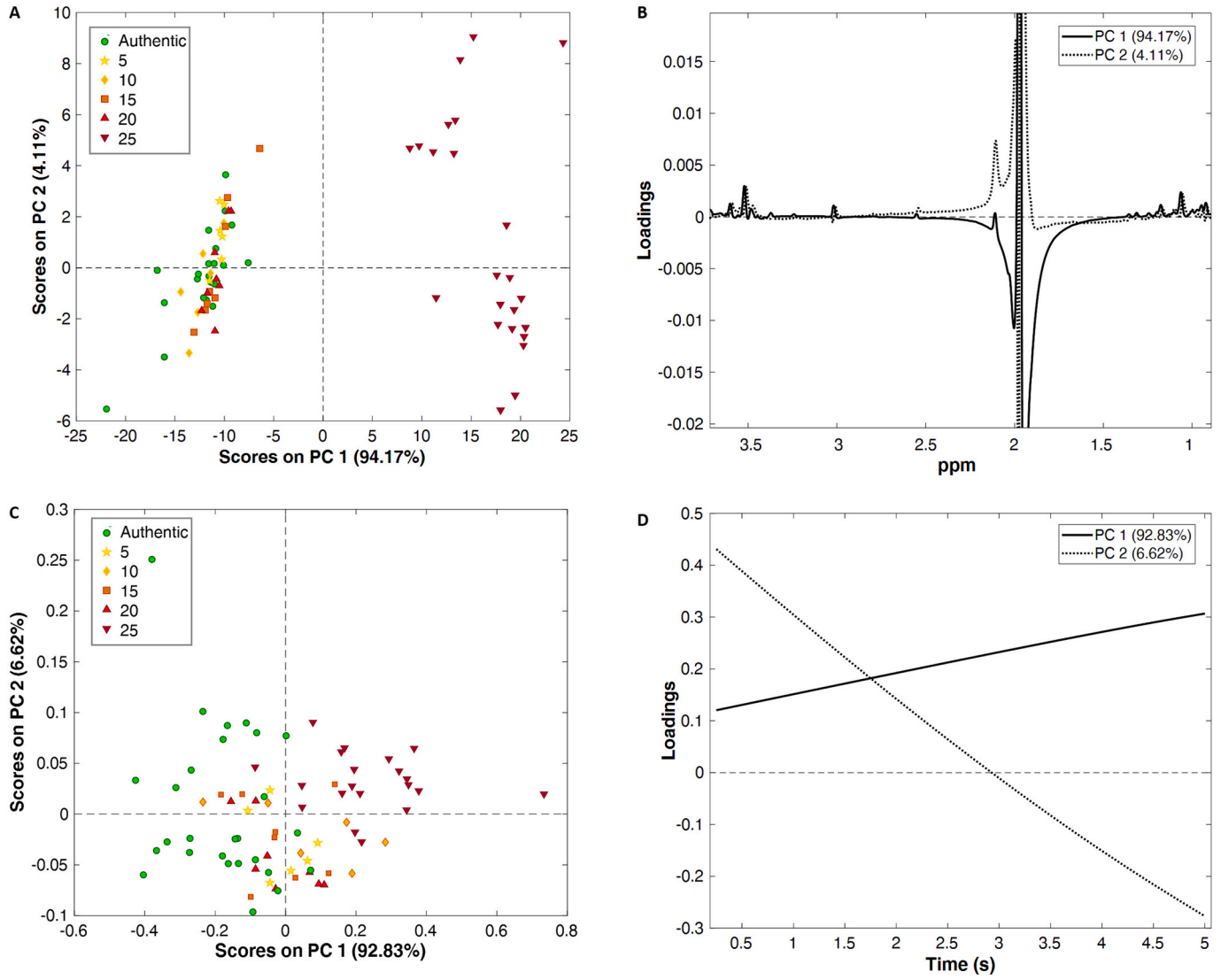

Fig. 5. Principal Component Analysis models developed with LF-NMR monodimensional spectra (A, score plot; B, loading plot) and acetic acid T2 (C, score plot; D, loading plot) datasets.

Table 1
Results of Linear Discriminant Analysis for discrimination of authentic and adulterated vinegars: correct classification percentages obtained with the 15 most informative variables selected for the aliphatic/alcoholic region from 3.75 to 0.90 ppm (region A), the region from 2.00 to 3.75 ppm (region B), and the region from 1.50 to 0.90 ppm (region C), as well as for the acetic acid and water T2 spin–spin relaxation times. For the spectral data, also the selected variables are indicated. N, number of samples in each class.

Dataset	Calibration		Cross-validation		Prediction	
	Authentic	Adulterated	Authentic	Adulterated	Authentic	Adulterated
N=	23	34	23	34	9	18
Range A	100.0	100.0	100.0	100.0	100.0	100.0
Selected variables (ppm)			1.751, 1.754, 1.755, 2.503, 2.504, 2.544, 2.571, 2.974, 2.992, 3.003, 3.461, 3.502, 3.636, 3.647, 3.664			
Range B	90.4	93.0	73.9	83.8	100.0	83.3
Selected variables (ppm)			3.364, 3.368, 3.373, 3.491, 3.664, 3.742, 3.744, 3.752, 3.753, 3.663, 3.667, 3.728, 3.734, 3.735, 3.745			
Range C	95.8	97.2	79.2	86.1	100.0	94.7
Selected variables (ppm)			0.919, 0.921, 0.926, 1.082, 1.149, 1.163, 1.239, 1.288, 1.302, 1.316, 1.320, 1.322, 1.362, 1.478, 1.499			
T2 water	78.3	85.3	56.5	73.5	66.7	55.6
T2 acetic acid	82.6	82.4	60.9	64.7	44.4	77.8

production methods (organic/conventional) to origin protection. In particular, great attention was paid to the protection of high-quality wine vinegars, such as protected designations of origin (PDOs). For example, Ríos-Reina, Callejón, Savorani, Amigo, and Cocchi (2019) obtained a reliable PLS-DA model to discriminate Spanish PDOs ("Vinagre de Jerez", "Vinagre de Condado de Huelva", and "Vinagre de Montilla-Moriles"). The 65 samples analyzed were divided by the Duplex algorithm into a calibration (47 samples) and a test (27 samples) set to build and validate the model, respectively. The robustness and reliability of the model, constructed with a small number of samples, was assured by repeating 5 times the splitting process. The weighted correct classification rate was higher than 90%. Similarly, different research groups developed models to discriminate Traditional Balsamic Vinegar of Modena and Balsamic Vinegar of Modena according to aging, obtaining sensitivity and specificity higher than 85% in cross-validation (Truzzi, Marchetti, Piazza, & Bertelli, 2023) or 80% correct classification in external prediction (Consonni et al., 2008). Also in this case the sample number was limited. From an initial pool of 72 vinegars of known age, 53 samples were selected by a D-optimal onion design, thus guaranteeing a closely resemble structure of the initial pool in terms of distribution of young, old, and very old samples.

More recently, Mascareli et al. (2023) developed PLS-DA on NMR data to classify vinegar according to raw materials and aging time. They obtained 100% accuracy in prediction when discriminating alcohol from fruit-based vinegars. However, those results were obtained by HF-NMR requiring sample preparation with D_2O and authentic vinegars, thus not considering possible adulterations. Before PLS-DA, samples of each class were split using the Kennard-Stone algorithm in training set (67% of samples) and test set (33% of samples).

Less research activity has been conducted to detect illegal adulteration practices (i.e., watering, sugaring, addition of synthetic acetic acid or table grapes) using NMR spectroscopy, especially in comparison with other techniques such as NIR (Ríos-Reina, Camiña, Callejón, & Azcarate, 2021). Boffo et al. (2009) developed KNN, SIMCA, and PLS-DA models from ^{1}H NMR spectra, obtaining a prediction ability of 100% when discriminating wine, apple, alcohol, and agrin authentic vinegars. Even though they reached a full prediction ability, they used a quite small number of samples (7 wine, 4 apple, 11 alcohol and agrin) to build the models and only 5 samples for the validation procedure, thus impairing the robustness of the proposed models.

Comparing the results obtained in the present work with the research activities reported in the literature, it is possible to postulate that the stated hypothesis can be confirmed. In particular, a high correct classification rate (up to 100% in prediction) was achieved with a reliable sampling design and rigorous data analysis for a feasibility study. The obtained results give to LF-NMR a relevant position in the field of vinegar adulteration among the other spectroscopic approaches (Cavdaroglu & Ozen, 2021) more prone to miniaturization (Grassi & Alamprese, 2024). Indeed, the present work demonstrated the feasibility to discriminate white wine vinegars from adulterated one by LF-NMR without sample pretreatment thank to the development of LDA models based on fifteen variables selected among the signals recorded in the range 3.75 to 0.90 ppm.

4. Conclusions

In conclusion, the study demonstrated that LF-NMR monodimensional spectra can be successfully used in LDA models to discriminate white wine vinegars from products adulterated with alcohol vinegars. The proposed method has the advantage to use the sample without pretreatment, thus resulting in a rapid and non-destructive untargeted approach. Moreover, the use of a LF-NMR spectrometer instead of a HF-one makes the method more approachable by food industries, being the instruments cheaper and more user-friendly.

The success of the proposed untargeted approach paves the way for the study of wider authentication models, based on a larger number of samples and including different possible adulterants. Wide and robust authentication models are indeed of great importance for food industries and control bodies, to fight commercial frauds in the sector of vinegar.

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CRediT authorship contribution statement

Silvia Grassi: Writing – review & editing, Writing – original draft, Visualization, Software, Methodology, Investigation, Data curation, Conceptualization. **Gigliola Borgonovo:** Writing – original draft, Formal analysis, Data curation. **Matteo Gennaro:** Writing – original draft, Formal analysis, Data curation. **Cristina Alamprese:** Writing – review & editing, Writing – original draft, Supervision, Resources, Project administration, Funding acquisition, Conceptualization.

Declaration of Generative AI and AI-assisted technologies in the writing process

During the preparation of this work the author(s) did not use any AI tool or service. The authors wrote, reviewed and edited all the text and take full responsibility for the content of the publication.

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.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.foodchem.2024.139953.

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