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Recent advances in NMR-based metabolomics of alcoholic beverages

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ABSTRACT

Alcoholic beverages have a complex chemistry that can be influenced by their alcoholic content, origin, fermentation process, additives, and contaminants. The complex composition of these beverages leave them susceptible to fraud, potentially compromising their authenticity, quality, and market value, thus increasing risks to consumers' health. In recent years, intensive studies have been carried out on alcoholic beverages using different analytical techniques to evaluate the authenticity, variety, age, and fermentation processes that were used. Among these techniques, NMR-based metabolomics holds promise in profiling the chemistry of alcoholic beverages, especially in Asia where metabolomics studies on alcoholic beverages remain limited.

Contents

1. Introduction

Asia has a long history of local production of alcoholic beverages dating back hundreds of years (Wiwanitkit, 2019). Despite the variety of alcoholic beverages in terms of taste, odor, and appearance, global production of alcohol is increasingly dominated by a shrinking number of companies (Day & McSweeney, 2016; Luo et al., 2020; Wardencki, 2018; Jernigan & Ross, 2020). In addition to being a substantial part of the food sector worldwide, production and consumption of alcoholic beverages play an important role in food security and health (Agnoli et al., 2018; Rugani, 2018).

The flavor and aroma of alcoholic beverages are attributed to their complex composition of chemical compounds such as esters, alcohols, acids, lactones, carbonyl compounds, acetals, phenols, and sulfur- and nitrogen-containing volatile compounds (Charapitsa et al., 2021; Uzhel et al., 2020; Zhou et al., 2015). This complexity means that alco-

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holic beverages are susceptible to adulteration, creating the challenging need for authentication. Falsification of labels or fraud in a commodity is considered a criminal offense (Neufeld & Rehm, 2018; Patra et al., 2019). Thus, proper labelling must accurately reflect a product's information in order to fulfill the expectations of consumers. The adulteration of alcoholic beverages is performed to increase the alcoholic strength of a product with cheaper sources of sugar and starch besides grapes or other fruits (Newman et al., 2018). Another adulteration practice involves watering down beverages to lessen the alcohol content of alcoholic beverages (Lachenmeier, 2016a, 2016b). Adulterations have been reported all over the world including incidents of unrecorded alcohol samples from China exceeding the specified alcohol content (Newman et al., 2018) and deaths in the Philippines due to suspected poisoning reportedly caused by *lambanog*, a Filipino alcoholic drink (Crisostomo, 2018; FDA, 2018a, 2018b; Lim, 2019).

Authenticity methods are in increasing demand for commercially fermented food and beverages, and much progress has been reported such as the verification of alcoholic beverage authenticity and detection of adulteration (Gliszczyńska-Świgło & Chmielewski, 2017; Pereira et al., 2018). Traditional methods such as sensory analysis for food authentication are still being used as an integral part of product standards and quality control (O'Sullivan, 2017; "Uses and good practices of sensory evaluation in the alcoholic beverage industry," 2017). One of the most recent and advanced analytical techniques that could improve the expensive and time-consuming methodologies is metabolomics–the comprehensive qualitative and quantitative analysis of the distribution of all metabolites present in a given biological system (Kueger et al., 2012; López-Ruiz et al., 2019). This type of –omics is employed to study the impact of agricultural practices, processing, and storage on the global chemical composition of food (García-Cañas & Simó, 2019; Zhang et al., 2020). In addition, this method is important for identifying novel bioactive compounds, authenticating product, and classifying region-of-origin (Johanningsmeier et al., 2016). Using the –omics studies, *foodomics* has been developed for investigating traditional and new issues in food analysis through the use of epigenomics, genomics, transcriptomics, proteomics, and metabolomics tools (Braconi et al., 2018; Cifuentes, 2012). Moreover, metabolomics has been successfully applied to display the metabolite profiles in various alcoholic beverages to assess safety, authenticity, quality, and traceability (Alañón et al., 2015). NMR-based metabolomics provides structural and quantitative information, which can be advantageous in the identification of unknown metabolites (Brennan, 2014). In general, the use of metabolomics in alcoholic beverages has created new opportunities to help monitor the production process for product safety, quality, and traceability (Hirst & Richter, 2016; Rizo et al., 2018; Singh et al., 2017). While alcoholic beverages have a high rate of adulteration amongst commodities, limited studies have been conducted for these beverages in Asia. This review focuses on the role of metabolomics applications in the profiling of alcoholic beverages in Asia, focusing on recent advances in NMR-based metabolomics approaches to ensure quality and safety in alcoholic beverages.

2. Metabolomics in wine and alcoholic beverages

Recent studies describing NMR-based metabolomics in alcoholic beverages show its growing field of applications. A brief comparison in terms of advantages, disadvantages, and compounds/properties analyzed using different analytical techniques are presented in Table 1. Significant emphasis is given to the implementation of the NMR techniques in the following section, and relevant and up-to-date examples of applications on alcoholic beverages are presented.

Nuclear magnetic resonance (NMR) spectroscopy allows identification of compounds without the need for complicated sample preparation. This spectroscopic technique can discriminate compounds that have identical masses and can be used to elucidate metabolic mechanisms and pathways using stable isotope labeling (Markley, 2017). In addition to NMR spectroscopy, analytical techniques such as gas chromatography (GC) or liquid chromatography (LC) and Fourier transform (FT) methods are also employed in wine metabolomics studies. Some of the separation techniques are used in tandem with mass spectrometry (MS) for its high selectivity and sensitivity as well as the potential to identify metabolites (Abd Ghafar et al., 2020; Karamanou & Aliferis, 2020). Combining a separation technique such as GC or LC with MS provides isobar separation, reduces the complexity of the mass spectra through metabolite separation in the time dimension, and delivers additional information on the physicochemi-

Table 1
Selected Representative Analytical Techniques used in Alcoholic Beverage Metabolomics.

Technique	Advantages	Disadvantages	Compounds/Properties Analyzed	References
LC–MS	High throughput and good coverage of metabolites Minimal sample pre-treatment or derivatization	Data may have biological or analytical variability due to sample preparation, instrument condition, or operating environment	Polyphenols, amino acids, biogenic amines, and ammonium ions, *trans*-resveratrol, allergenic residues, characterization of whiskey and beer	Donato et al., 2016; Lu, Liao, & Chen, 2018; Redruello et al., 2017; Spinelli et al., 2019; Collins et al., 2014; Hughey et al., 2016
GC–MS	High sensitivity, peak resolution, and reproducibility	Requires sample derivatization or pre-treatment due to differences in polarity of the analytes and to enhance the volatility and thermal stability of the metabolites Extensive treatment of the sample may result in the loss of volatile metabolites	Ethanol, biogenic amines, semi-volatile and volatile constituents	Cai, Rice, Koziel, & Dharmadhikari, 2017
FT–IR	Can analyze most samples Relatively inexpensive Fast and simple analysis Nondestructive	Difficulty in analyzing molecules that do not vibrate Trouble differentiating compositions in a mixture	Ethanol, grape quality, total antioxidant capacity, wine polysaccharides, quality control	Barnaba, Bellincontro, & Mencarelli, 2014; Boulet, Williams, & Doco, 2007; Lachenmeier, Richling, López, Frank, & Schreier, 2005; Versari, Parpinello, Scazzina, & Rio, 2010
NMR	Nondestructive Fast analysis time Requires minimal sample preparation Robust Reproducible	Difficulty in detecting metabolites at very low concentrations, thus requiring solvent suppression methods to improve resolution for better metabolite identification	Structural characterization, classification of variety and vintage, authentication, quality control	Fan et al., 2018; Geana et al., 2016; Kew, Goodall, & Uhrín, 2019; Monakhova, Kuballa, & Lachenmeier, 2012; Fotakis & Zervou, 2016; Cassino et al., 2019; Belmonte-Sánchez et al., 2020; Mannu et al., 2020; Gougeon et al., 2019; Sánchez-Estébanez et al., 2018; Dzhimak et al., 2017; Ciepielowski et al., 2019

cal properties of the metabolites (Dettmer et al., 2007; Zeki et al., 2020). Only a brief discussion of non-NMR techniques is presented in this paper. We refer the readers to more comprehensive reviews that cover the role of MS, gas or liquid chromatography, and FT methods in metabolomics (Aszyk et al., 2018; Beale et al., 2018; Esteki et al., 2018; Hird et al., 2014).

2.1. Mass spectrometry (MS)

Mass spectrometry is one of the common analytical techniques for the analysis of metabolites in complex biological samples. Its high sensitivity and fast data acquisition enable MS to be one of the most widely used analytical tools in the field of metabolomics (Brunius et al., 2016). Liquid chromatography and gas chromatography are separation methods typically combined with MS. Each of these methods provide solutions to complex mixture analyses and have been used extensively in metabolomics. LC coupled with electrospray-ionization MS (LC–ESI-MS) is commonly applied for detecting metabolites in complex biological samples and in profiling unknown metabolites. This "soft" ionization method forms intact molecular ions and aids in initial identification (Aszyk et al., 2018; Bowen & Northen, 2010), allowing for a wider range of metabolite detection than GC–MS because metabolite volatility is not required (Vinaixa et al., 2016). In addition, good chromatographic separation will result in higher quality MS data with reduced background noise.

Recent applications of LC–MS in alcoholic beverages include analysis of polyphenolic compounds, biogenic amines, resveratrol, and allergenic residues (Donato et al., 2016; Lu et al., 2018; Redruello et al., 2017; Spinelli et al., 2019). Successful identification of metabolites requires accurate mass measurements and high-quality mass spectra. By obtaining the mass spectra, the structures of the metabolites of interest can be further confirmed, as fragmentation helps in deducing possible structures of metabolites during spectra interpretation (Tada et al., 2019). Because fragmentation patterns may contain a wide variety of possible metabolite structures, it is difficult to identify metabolites through MS alone. To solve this problem, the detected ions can be identified using a spectral library match or mass-based search using the retention index derived from retention time (Matyushin et al., 2020). However, no such index currently exists for LC–MS experiments, and the comparison of retention times can only be performed under identical experimental conditions (Chaleckis et al., 2019). On the other hand, use of collisional cross section (CCS) values from LC–MS coupled with ion mobility spectrometry (IMS) can give additional confidence in identifying detected ions (D'Atri et al., 2018; Nichols et al., 2018).

Similar to LC–MS, GC–MS has been widely used in the food sector and in wine metabolomics studies due to its high sensitivity, peak resolution, and reproducibility (Kang et al., 2016; Korban et al., 2021; Stupak et al., 2017). However, one drawback of GC–MS is the complex sample preparation prior to analysis, which aims to produce extracts that are compatible with the GC technique. Pre-treatment and extraction of the sample are sometimes required to enhance the volatility and thermal stability of the metabolites. Sample derivatization is one of the pretreatment procedures commonly used in GC–MS analysis of metabolites to modify polarity and facilitate chromatographic separation on a column of low polarity (Moros et al., 2017). One-step reactions are preferred, but sometimes, two-step derivatizations are more desirable when the obtained derivatives show better chromatographic separation or more adequate mass fragmentation (Miyagawa & Bamba, 2019).

GC–MS analysis also requires an extraction process in order to separate the metabolites and enhance their concentration. Extraction of metabolites from the sample should be as comprehensive as possible, but care should be observed to prevent metabolite degradation or modification (Desbrosses et al., 2005; Pinu et al., 2017). Recent methods have focused on improving the extraction of metabolites from alcoholic beverages using solid phase microextraction (SPME) (Cai et al., 2017; Papageorgiou et al., 2018; Riu-Aumatell et al., 2014; Sagandykova et al., 2017), or stir bar sorptive extraction (SBSE) (Hevia et al., 2016; Takase et al., 2015; Zhou et al., 2015) methods. Most analyses using GC–MS involve determination of volatile aromatic compounds, organic acids, amino acids, carbohydrates, and polyphenols in wines and alcoholic beverages (Cai et al., 2017; Hevia et al., 2016; Papageorgiou et al., 2018; Takase et al., 2015).

2.2. Fourier transform infrared (FT-IR) spectroscopy

In contrast with the chromatographic techniques previously discussed, FT-IR spectroscopy provides rapid and nondestructive analysis of alcoholic beverages with little sample preparation required. It is a valuable metabolic fingerprinting tool that has the ability to simultaneously analyze compounds such as carbohydrates, polyphenols, amino acids, proteins, and polysaccharides (Anjos et al., 2020; Garcia-Hernandez et al., 2020; Puxeu et al., 2019; Teixeira dos Santos et al., 2016) and has been applied for quality control and authentication (Anjos et al., 2016; Basalekou et al., 2020; Yadav & Sharma, 2019). However, its main drawback is its inability in the differentiation of the chemical constituents present in a mixture (Kumar et al., 2019; Lerma-García et al., 2018). Nevertheless, combining FT-IR spectroscopy with different analytical techniques provides information on elemental composition, molecular structure, and chemical speciation. This enables accurate identification of the main compounds found in alcoholic beverages such as ethanol, sugars (e.g., glucose and fructose), and organic acids (Li et al., 2014; Wang, Shao, Chen, Meng, & Li, 2019; Wang et al., 2017).

2.3. NMR spectroscopy

Nuclear magnetic resonance (NMR) spectroscopy is a widely used analytical tool in metabolomics due to its non-destructive nature and rapid analysis (Wishart, 2019), providing a high-throughput method with minimal sample preparation, an advantage over chromatographic techniques. Unlike FT-IR spectroscopy, NMR-based techniques enable simultaneous identification and absolute quantification of compounds; prediction of authenticity parameters such as variety, geographical origin and vintage; and the potential for the detection of unexpected and unknown fraud or infection (Trimigno et al., 2015). NMR is widely used today for alcoholic beverage analysis, with continuous novel research for additional uses. (Diamantidou et al., 2018; Fan et al., 2018; Geana et al., 2016; Kew et al., 2019; Kuballa et al., 2018).

Due to proton abundance in metabolites, proton (^{1}H) NMR spectroscopy is often used for profiling an extensive range of metabolites such as amino acids, polyphenols, sugars, and other organic compounds present in alcoholic beverage samples (Bordiga et al., 2016; Lu et al., 2018; Peterson & Waterhouse, 2016; Zerbib et al., 2018). In particular, ^{1}H NMR methodologies have been applied to determine chemical constituents (phenolics, sugars, and organic acids) and to measure the alcohol content for authenticating the quality of alcoholic beverages (Belmonte-Sánchez et al., 2020; Polak & Bartoszek, 2015). The chemical composition obtained from NMR spectroscopic data constitutes the so-called alcohol beverage metabolome, which can vary significantly depending on the geographic origin, variety, aging, fermentation process, and quality of alcoholic beverages (Alañón et al., 2015; Bokulich et al., 2016; Dzhimak et al., 2017; Spraul et al., 2015). Because metabolites from a sample may produce complex 1D NMR spectra with poorly resolved proton resonances, high-field NMR instruments and two-dimensional NMR (2D NMR) spectroscopy are typically employed to improve resolution and provide additional NMR spectral data to aid in the interpretation of NMR data (Bo et al., 2019; Moser et al., 2017).

High-resolution NMR spectroscopy has become the most suitable technique for analyzing beverages at the molecular level following the development of high-field NMR spectrometers. Multivariate statis-

tical analysis or chemometrics in combination with NMR spectroscopy for data processing have also improved the sample analysis by reducing analysis time and providing more informative results (Medina et al., 2019). Chemometric methods such as partial least squares (PLS), principal component analysis (PCA), and partial least squares-discriminant analysis (PLS-DA) can determine characteristic patterns of compounds or parameters related to a geographical origin, the adulteration of samples, or certain specific conditions such as processes, storage, climate, and variety (Borràs et al., 2015; Magdas et al., 2019).

While the ultimate goal of metabolomics is the identification and quantitation of all small molecules in an alcoholic beverage sample, a single method allowing complete metabolome analysis does not exist. Several complementary analytical techniques should be applied to improve the scope and identification ability in metabolite analysis.

3. Recent applications of NMR-based metabolomics

During the last few years, NMR-based metabolomics has been applied to characterize complex food matrices and has been widely used in profiling food samples for quality and authentication assessment (Rocha & Sheen, 2018). Authenticity of alcoholic beverages has been extensively investigated because it is a product that can be easily adulterated due to its signature chemical properties (high alcohol content, low pH) and its availability throughout the world (Ahmad et al., 2018; Cioch-Skoneczny et al., 2020). In addition to authenticity, traceability has an important role in the quality assurance of alcoholic beverages. A traceability protocol must be established on the supply chain to track and manage all representative events which may arise from raw material production to selling the final product (Kamiloglu, 2019). Alcoholic beverages are valuable commodities that are recognized for their geographical region, variety, and age. These factors are the main determinant for determining consumer preference and market value (Anderson et al., 2019). Therefore, the use of a metabolomics platform adds value by providing fingerprints to classify samples of alcoholic beverages by variety, age, fermentation processes, and geographic origin. The following section discusses how NMR-based metabolomics have been applied to evaluating different alcoholic beverages in Asia.

3.1. Variety effect

Different kinds of plant and fruit sources are needed to create an alcoholic beverage. The final product is both complex and information-intensive, so it is important to understand the behaviors and characteristics of different varieties (Chay et al., 2017; Li & Sun, 2019) from the plant source to the finished product.

The characteristic metabolites of Cabernet Sauvignon and Shiraz from Shanxi province, China were analyzed by ^{1}H NMR spectroscopy

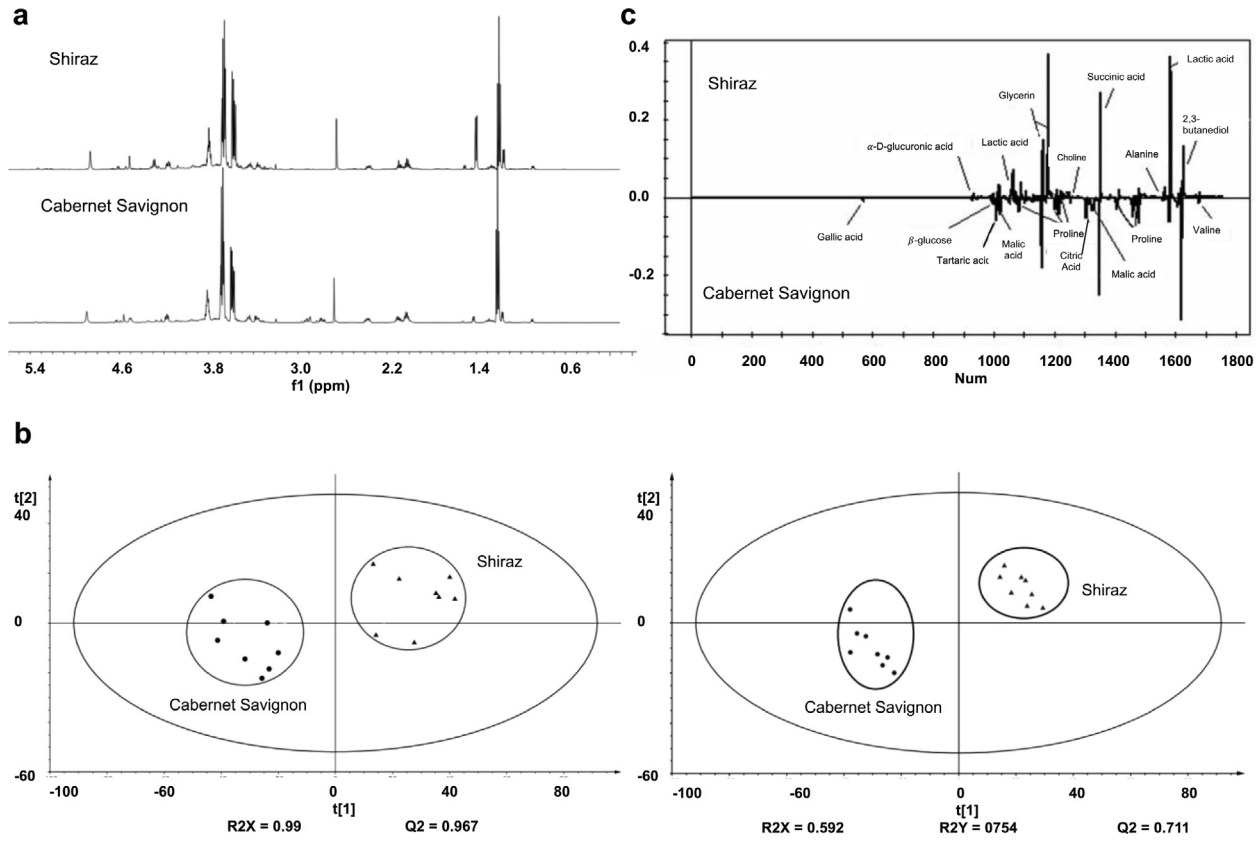

Fig. 1. Panel 1a shows the ^{1}H NMR spectrum of Cabernet Sauvignon and Shiraz dry red wines. Panel 1b (left) shows the PCA score plot based on the ^{1}H NMR spectra of Cabernet Sauvignon and Shiraz dry red wines. Based from the score plot, there was a significant distinction between Cabernet Sauvignon and Shiraz dry red wine, showing a significant difference of the metabolites from the two wine samples. The cumulative contribution rate, $R^2X = 0.99$, and $Q^2 = 0.967$, indicates that the established PCA model was of good quality. Panel 1b (right) shows the PLS-DA score plot based on the ^{1}H NMR spectra of Cabernet Sauvignon and Shiraz dry red wines. The cumulative contribution rate from PLS-DA was $R^2X = 0.592$, $R^2Y = 0.754$ and $Q^2 = 0.711$, which are all greater than 0.5, indicating that the model is valid. Based from the PLS-DA score plot, the distinction between Cabernet Sauvignon and Shiraz dry red wines was more pronounced than PCA score plot. Panel 1c shows the PLS-DA loading plot based on the ^{1}H NMR spectra of Cabernet Sauvignon and Shiraz dry red wines. The higher peak in the loading plot indicated that content of the corresponding metabolite was higher in dry red wine, and the lower one indicated that content of the corresponding metabolite was lower. Reprinted (adapted) with permission from Zhu, Hu, Lu, and Xu (2018). Analysis of Metabolites in Cabernet Sauvignon and Shiraz Dry Red Wines from Shanxi by ^{1}H NMR Spectroscopy Combined with Pattern Recognition Analysis, Open Chemistry, 16(1), 446–452. doi: https://doi.org/10.1515/chem-2018–0052).

(Zhu et al., 2018), and the biomarkers for the differentiation between wine samples were identified using pattern recognition analysis. The NMR spectra (Fig. 1a) showed the proton signals of the major metabolites of Cabernet Sauvignon and Shiraz red wines. The PCA score plot (Fig. 1b, left) from the ^{1}H NMR data of Cabernet Sauvignon and Shiraz dry red wines suggested significant differences in metabolites from these two wines. Data from the PLS-DA score plot (Fig. 1b, right) showed more pronounced differences in metabolites present between Cabernet Sauvignon and Shiraz dry red wines compared to the one illustrated in PCA score plot. The main contributors to these metabolite differences as identified from the PLS-DA loading plot (Fig. 1c) were proline, tartaric acid, glycerin, lactic acid, choline, succinic acid, and gallic acid. Proline in particular had largest difference in content (0.268 g/L difference) between the two wine samples. The proline content had a direct impact on the depth and complexity of wine taste and flavor, affecting both the sweetness and bitterness in the wine (Zhu et al., 2018). Because proline in wines is not consumed by fermentation and maturation, the difference in proline content in the study is primarily attributed to the different wine varietals (Zhu et al., 2018). The PLS-DA score plot also revealed significant difference in tartaric acid content (0.022 g/L) between the two wine samples. This nonvolatile acid could impart dark red color to wines, and its content depends on the type of grape that is used in making wine (Zhu et al., 2018). Finally, the difference in succinic acid content (4×10^{-3} g/L) between the two samples was also found to be significant. Succinic acid content is dependent on yeast nitrogen metabolism and had an influence on the sweetness and body thickness of wines (Zhu et al., 2018). Overall, these metabolite differences may be used for discrimination between Cabernet Sauvignon and Shiraz dry red wine samples. Further, the significant differences in metabolite content between these wines could be utilized for establishing an evaluation system for assessing the quality and authenticity of these wines.

Another study investigated the metabolites present in different varieties of dry red wine (Cabernet Sauvignon, Merlot, and Cabernet Gernischt) vinified in Changli, Hebei province, China, using ^{1}H-based techniques (Hu et al., 2020). NMR data was processed by integrating the chemical shift between 0 and 10 ppm followed by removal of the internal standard (4,4-dimethyl-4-silapentanesulfonate, DSS), residual ethanol, and residual water peaks. The integral data was subsequently normalized and subjected to multivariate statistical analysis. While most of the metabolites (mainly amino acids, organic acids, sugar, phenolic compounds, etc.) that were identified from these wine samples are similar, their concentrations are different. PLS-DA loading plots of the three dry red wines revealed content differences in proline, ethyl acetate, valine, 2,3-butanediol, succinic acid, glycerol, lactic acid, β-glucose, malic acid, tartaric acid, α-D-glucuronic acid, choline, alanine, and gallic acid. Relative contents of these metabolites were determined by comparing the peak area of their proton resonances to that of DSS. Among these metabolites, succinic acid and glycerol contents were highest in Merlot. On the other hand, gallic acid content was highest for Cabernet Sauvignon. Lactic acid, alanine, proline, and malic acid contents were highest in Cabernet Gernischt. These main metabolites were closely related to the flavor, taste, and functional activity of wine. For example, glycerol contributes to mouth-feel perception because of its sweet taste and viscous nature while succinic acid makes wines rich in flavor and mellow in taste. Succinic acid content is influenced by malic acid levels and yeast species (Hu et al., 2020). Because the three dry red wines underwent the same brewing process, the differences in their glycerol and succinic acid contents are most likely due to the wine varietals. The other main metabolites namely gallic and malic acids have reported health benefits. This study provided an effective and feasible method for establishing metabolic content profiles for dry red wines that can be used for the identification of dry red wine varieties and could offer reasonable advice for consumers to choose suitable dry red wines according to their preferences (i.e., flavor, taste, and health).

3.2. Aging/vintage effect

The changes in alcoholic beverages after fermentation and before bottling are also essential in their metabolomics. Aging is a result of oxidation and non-oxidative processes (Li et al., 2017). To ensure consistent aging before bottling, beer, wine, and spirits undergo “maturation” – (Miller & Miller, 2019) a long and expensive process in which the beverage acquires a distinct flavor due to factors such as type of alcohol and wood matrix (González-Robles & Cook, 2016; Morishima et al., 2019). The effect of aging on alcoholic beverages is monitored by the changes in their composition such as their anthocyanin content (Li et al., 2017), yeast interactions (Legras et al., 2016), and aging techniques (Santos et al., 2019).

^{1}H NMR analysis coupled with multivariate statistical analyses (PCA and PLS-DA) and hierarchical cluster analysis (HCA) was used to investigate the genetic relatedness of grapevine (*V. vinifera*) cultivars grown on the same site and vinified through similar processes from Hebei, China (Hu, Yue, Zhu, Wen, & Zhang, 2015). NMR data was acquired using ^{1}H and ^{1}H–^{1}H correlation spectroscopy (COSY) NMR techniques and processed by integrating the chemical shift between 0 and 10 ppm followed by removal of DSS, residual ethanol, and residual water peaks. PCA and PLS-DA revealed discriminant grape metabolites such as proline, valine, tartaric acid, glucose, and total phenols (mostly gallic acid). Content of phenolic compounds are influenced by several factors such as grape cultivar, fermentation processes, maturation with wood contact, and wine age. While NMR assignments for these compounds are typically confounded by their complex structures, the total content of phenolic compounds can be a useful biomarker to discriminate wine in terms of grape cultivars. Hu et al. identified gallic acid as the main phenolic compound present at different levels in the tested wine samples: Ruby Cabernet > Cabernet Sauvignon > Merlot > Syrah > Zinfandel. PCA and PLS-DA also revealed several discriminating compounds arising from alcoholic fermentation: 2,3-butanediol, glycerol, valine and succinic acid. Overall, the NMR data coupled with multivariate and hierarchical cluster analyses presented groupings that reflect the genetic heritage of the cultivars and revealed important biomarkers for fermentation processes and cultivars discrimination.

Metabolite differences in 2009, 2010, 2011, and 2012 vintages of Cabernet Sauvingnon wines from Ningxia was explored using ^{1}H NMR techniques coupled with pattern recognition methods (PCA and PLS-DA) (Hu et al., 2016). The variation in metabolite compositions was found to be small while the differences in metabolite levels were found to be significant among the vintages (Fig. 2 showing discrimination of 2011 and 2012 vintages). The metabolites responsible for the differentiation were identified as 2,3-butanediol, ethyl acetate, valine, proline, succinic acid, lactate, acetic acid, glycerol, gallic acid and choline. Because fermentation technique, yeast, and grape varieties were similar for these wine vintages, varying concentrations of most of the discriminating metabolites were likely influenced by climatic factors such as average temperature, rainfall, and evaporation. For example, the higher-than-average temperature (17.65 °C) and evaporation (1562.1 mm) and less rainfall (243.5 mm) in 2009 were favorable climatic conditions for grapes to reach optimum ripeness, thus likely affecting their sugar content. This climatic condition in 2009 may have influenced the higher valine, 2,3-butanediol, gallic acid, and proline content in 2009 vintage relative to 2011 vintage, when the climatic conditions were characterized by lower-than-average temperature (15.24 °C) and evaporation (1423.6 mm) and high rainfall (262.2 mm). Taken together, this study opens new opportunities for exploring other discriminants that can be useful for classifying alcoholic beverages according to their vintage, especially in regions with the most variable climates.

3.3. Fermentation process

Alcoholic beverages are obtained through the process of fermentation of a sugar to ethyl alcohol and other minor products such as amino

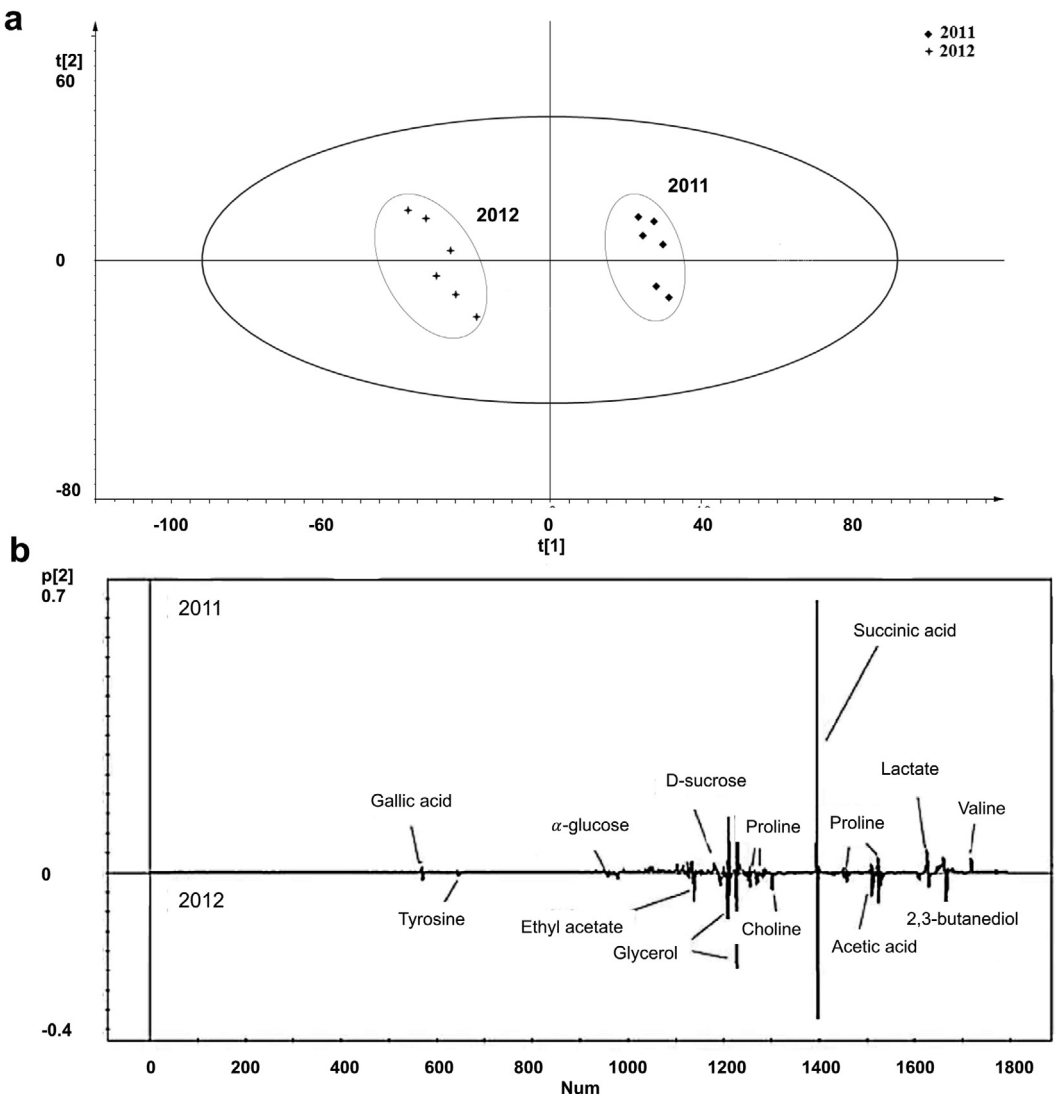

Fig. 2. Panel 2a shows the PLS-DA score plot chart from ^{1}H NMR spectra of 2011 and 2012 Chinese vintage Cabernet Sauvignon wines, while panel 2b shows PLS-DA loading plot chart from ^{1}H NMR spectra of 2011 and 2012 Chinese vintage Cabernet Sauvignon wines. The PLS-DA score plot showed clear separation between the 2011 and 2012 Chinese vintage Cabernet Sauvignon wines based on the first component, and the corresponding loading plot showed relatively high-load levels of valine, lactic acid, and succinic acid, with low levels of 2,3-butanediol, proline, acetic acid, choline, glycerol, D-sucrose, acetate, α-glucose, gallic acid, and tyrosine in the 2011 vintages, compared with the 2012 vintages. Reprinted (adapted) with permission from Hu, B., Zhao, Q., Yue, Y., Zhu, J., Lu, G., Li, H., … & Hardie, W. J. (2016). ^{1}H Nuclear magnetic resonance-based metabolomic study for Cabernet Sauvignon wines in different vintages (No. e2332v1). PeerJ Preprints.

acids and carbohydrates (Yıldırım, 2021). Most fermentations of alcoholic beverages are accomplished using strains of the yeast *Saccharomyces cerevisiae* (Walker & Stewart, 2016). The fermentation process is one of the most important steps in producing wine and other alcoholic beverages, and the process must be monitored in order to achieve a high quality product.

NMR-based metabolomics studies have also been applied in recent years to monitor the fermentation process of alcoholic beverages. For example, selected bacterial and yeast strains (*Bacillus licheniformis, Pediococcus pentosaceus, Lactobacillus plantarum, Pichia kudriavzevii, Wickerhamomyces anomalus, Saccharomyces cerevisiae, and Saccharomycopsis fibuligera*) were investigated for their amylolytic activity, ethanol tolerance, and metabolite production during sorghum-based laboratory-scale alcoholic fermentation using ^{1}H NMR spectroscopy coupled with multivariate statistical analysis (Li et al., 2018). Principal component analysis showed variations in the metabolites produced by different mixtures of pure cultures. *S. cerevisiae* was found to be associated with significant production of lactic acid (17.7 mmol/g with *S. cerevisiae* vs 0.3 mmol/g without *S. cerevisiae*). However, *S. cerevisiae* cannot convert starch into ethanol whereas *S. fibuligera* and *B. licheniformis* can convert starch or polysaccharides to soluble sugars such as maltose, maltotriose, and dextrin (Li et al., 2018). Therefore, *S. fibuligera* is introduced with *S. cerevisiae* in the alcoholic samples during fermentation. Lactic acid bacteria had high amylolytic and proteolytic activities, thus contributing to increased saccharification and protein degradation. Finally, *W. anomalus* was found to have a positive effect on the flavor of the *Daqu*-derived product as this species produces acetate esters such as ethyl acetate.

In recent years, inactive yeasts or deactivated yeasts are introduced in wine fermentation as they are reported to increase the rate of alcohol fermentation and influence the aroma components in wine. The effect of five inactive yeasts (Optim-White®, Noblesse®, Opti-LEES, Boster Blanchc®, and Mannostab®) on the metabolites of Chardonnay dry white wines vinified in 2016 in Hebei province, China was investigated using ^{1}H NMR spectroscopy coupled with multivariate analysis (PCA/PLS-DA) (Hu et al., 2019). The main metabolites of the wine

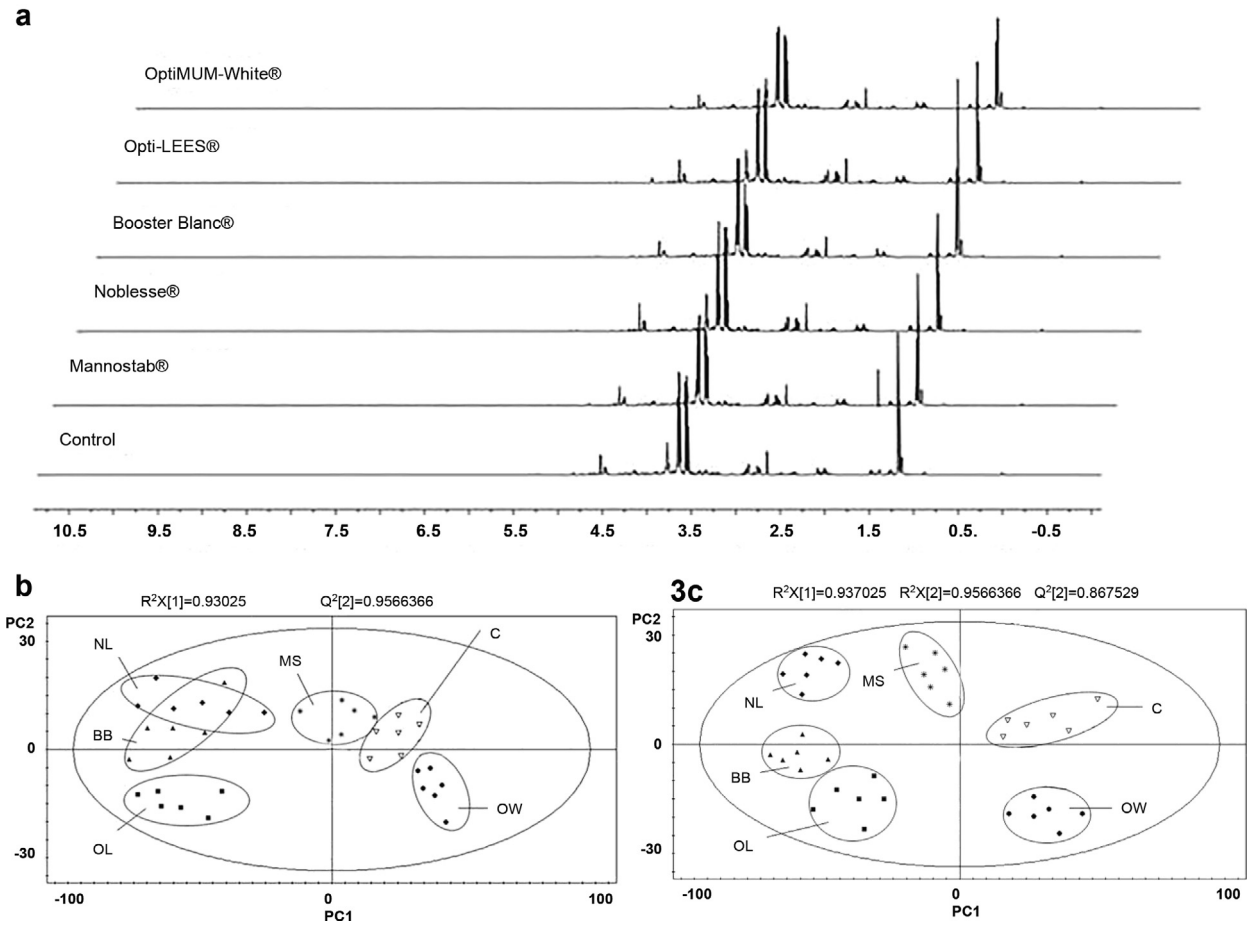

Fig. 3. Panel 3a shows the ^{1}H NMR spectra of all dry white wine samples. Most of the metabolites in Chardonnay dry white wine are concentrated in the range of 2.0–9.0 ppm, and the metabolites in the range of δ0.0–5.0 ppm are relatively dense showing that there were many kinds of metabolites in this interval and the content was relatively high. Panel 3b shows the PCA score plot based on the data obtained from the Chardonnay dry white wine. The difference between Chardonnay wine and sample wine with the inactive yeast could be distinguished, and each yeast addition group also has varying degrees of dispersion. $R^2X = 0.937$, $Q^2 = 0.957$ which indicates that the established PCA model is reliable. Panel 3c shows the PLS-DA score plot to further demonstrate between the wine samples. The $R^2X = 0.937$, $R^2Y = 0.957$, and $Q^2 = 0.867$, all of which were above 0.5, indicating that the model of the construction has a high quality. There is also a discrete relationship between the various wine samples as shown from the score plot. *(OW, OptiMUM-White®; OL, Opti-LEES; BB, Booster Blanc®; NL, Noblesse®; MS, Mannostab®; C, control)*. Reprinted (adapted) with permission from Hu, B., Cao, Y., Zhu, J., Xu, W., & Wu, W. (2019). Analysis of metabolites in chardonnay dry white wine with various inactive yeasts by ^{1}H NMR spectroscopy combined with pattern recognition analysis. AMB Express, 9(1), 140. https://doi.org/10.1186/s13568-019–0861-y.

samples were identified from NMR data (Fig. 3a), and the differences of metabolites between the wines with inactive yeast and the original wine were analyzed using PCA (Fig. 3b) and PLS-DA (Fig. 3c). PCA and PLS-DA plots revealed significant influence of the added inactive yeasts on the levels of polyols, organic acids, amino acids, and other metabolites in the dry white wine samples. The main metabolites were found to be glycerol, 2,3-butanediol, lactic acid, malic acid, tartaric acid, succinic acid, gallic acid, proline, alanine, valine, ethyl acetate and choline. The higher glycerol levels in Opti-LEES, Boster Blanchc®, and Noblesse® relative to the original wine suggest yeast-induced enhancement in activity and fermentation efficiency. Malic acid content is related to malolactic fermentation in which lactic acid bacteria convert maleate and citrate into lactate and other components (Hu et al., 2019). Higher levels of this organic acid in Optim-White® and Noblesse® relative to the original wine indicate the significant contribution of these inactive yeasts to the efficacy of malolactic acid fermentation. The contents of main amino acids valine, proline, and alanine in Noblesse® and Mannostab® were higher than that of the original wine. Other distinct metabolites such as ethyl acetate were also found to be higher in Opti-LEES, Boster Blanchc®, Noblesse®, and Mannostab® compared to that of the original dry white wine. In summary, this study provided a theoretical basis for using NMR-based metabolomics in monitoring composition and content differences among original dry white wine samples to investigate the effect of inactive yeast to fermentation process.

3.4. Quality control/authentication

Quality assessment and authentication are crucial issues to the food and beverage industry, especially with alcoholic beverages. The quality of alcoholic beverages is often assessed by physicochemical and sensory tests (Ickes & Cadwallader, 2017; Palczak et al., 2019). Physicochemical laboratory tests include routine analysis of density, alcohol, or pH values, while sensory evaluation relies on human experts. There is an interrelated role of sensory, emotional, and cognitive responses, which affects the panelists' issues of personal taste (Palczak et al., 2019; Stelick & Dando, 2018).

In recent years, integrated approaches using metabolomics in addition to sensory properties to assess the quality of alcoholic beverages have shown promise to improve upon sensory analysis (Nikolantonaki et al., 2018; Taniguchi et al., 2019). The assessment of alcoholic beverage authenticity is receiving increased attention

since alcoholic beverages are often subject to fraudulent practices (Lachenmeier, 2016a, 2016b; Rodgers, 2020). The adulteration of such commodities consists of the addition of any substance to the natural wine to change its composition. The most common forms of adulteration include addition of water and sugar, mixing with lower quality wines, and fabricating labels (Lachenmeier, 2016a, 2016b; Zhang & Xue, 2016). Adulteration of alcoholic beverages is especially concerning due to the potential to harm customer health. For example, recent samples of *lambanog*, a popular alcoholic beverage from the Philippines, tested positive for high levels of methanol. The *lambanog* products tested by the Food and Drug Administration of the Philippines (FDA) were found to have methanol levels ranging from 6.5 percent to 21.8 percent. These levels greatly exceeded the Philippine National Standard and is suspected to have caused the deaths of some individuals (Crisostomo, 2018; FDA, 2018a, 2018b; Lim, 2019). Thorough chemical analysis of the metabolic composition of such products could prevent these frauds. However, the association between chemical composition and wine origin is a challenge due to variability in the chemical composition and the chemical changes produced during the winemaking and fermentation stages. These issues highlight the need for modern analytical techniques and advanced data analysis to obtain reliable information to ensure the authenticity of wines.

4. Conclusion

The complex chemistry of alcoholic beverages and the factors influencing its chemical composition make it a difficult task to characterize such samples. Alcoholic beverage quality and authenticity need to be monitored from the plant source to the finished product, making profiling every step in the production a challenging task. Metabolomics is an emerging field of –omics research. NMR spectroscopy contributes greatly to metabolomics by producing information-rich, reliable, and reproducible data sets in both non-targeted or global and multivariate statistical analysis. Recent studies have demonstrated the promising potential of NMR-based metabolomics in understanding wine chemistry and profiling metabolites in alcoholic beverages.

New developments in NMR hardware and techniques, especially in combination with chemometric methods, have advanced detection of various metabolites in terms of geographical origin, variety, vintage, fermentation, and quality assessment in wines and alcoholic beverages. NMR-based metabolomics studies in Asia have exhibited promising opportunities for determining biomarkers and for establishing metabolomic “fingerprint” that may have important implications for assessing the quality, authenticity, and safety of alcoholic beverages. For example, phenolic compounds may be used as biomarkers for determining wine varietals. However, assignment of proton signals is limited by their complex structures; therefore, the use of other NMR-based techniques such as DOSY ^{1}H NMR may help in identifying this class of compounds in alcoholic beverages. DOSY ^{1}H NMR is also useful for providing information on the relative molecular sizes of the metabolites (e.g., maltose and maltotriose versus glucose and fructose) present.

Further research should be developed using multidimensional or orthogonal techniques, such as GC × NMR, or LC × NMR in metabolomics studies in the near future. Multidimensional methods should also be employed to enhance resolution of complex mixtures, achieve a higher peak capacity, and increase the sensitivity and selectivity. Investigation into novel applications using portable analysis may yield advantages such as decreased consumption of reagents and samples and fast automated analysis that can generate results in real time at a low cost (Spyros, 2016). Studies involving other alcoholic beverages aside from wine are seldom researched, especially in Asian countries; thus, it is recommended to turn these methods to locally-produced alcoholic beverages such as beer, whiskey, and those produced from other plant sources such as rice and coconut wine. In summary, research in metabolomics have enhanced our understanding of alcoholic beverages, but there is great potential for further advances both in methods used and their application to unraveling the components of complex matrices such as pharmaceuticals, dietary supplements, foods, and biological extracts.

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.

References

Abd Ghafar, S. Z., Mediani, A., Maulidiani, M., Rudiyanto, R., Mohd Ghazali, H., Ramli, N. S., & Abas, F. (2020). Complementary NMR- and MS-based metabolomics approaches reveal the correlations of phytochemicals and biological activities in Phyllanthus acidus leaf extracts. *Food Research International*. https://doi.org/10.1016/j.foodres.2020.109312.

Agnoli, L., Boeri, M., Scarpa, R., Capitello, R., & Begalli, D. (2018). Behavioural patterns in Mediterranean-style drinking: Generation Y preferences in alcoholic beverage consumption. *Journal of Behavioral and Experimental Economics*. https://doi.org/10.1016/j.socec.2018.06.001.

Ahmad, I., Ahmed, S., Yadav, Sah, P. P., Alam, & Zzaman. (2018). Study on the physicochemical, microbial and sensory characteristics of alcoholic beverage produced by indigenous method. In International Food Research Journal.

Alañón, M. E., Pérez-Coello, M. S., & Marina, M. L. (2015). Wine science in the metabolomics era. *TrAC - Trends in Analytical Chemistry*. https://doi.org/10.1016/j.trac.2015.05.006.

Anderson, K., Meloni, G., & Swinnen, J. (2019). Global alcohol markets: Evolving consumption patterns, regulations, and industrial organizations. *World Scientific Studies in International Economics*. https://doi.org/10.1142/9789811202094_0026.

Anjos, O., Comesaña, M. M., Caldeira, I., Pedro, S. I., Oller, P. E., & Canas, S. (2020). Application of functional data analysis and FTIR-ATR spectroscopy to discriminatewine spirits ageing technologies. *Mathematics*. https://doi.org/10.3390/MATH8060896.

Anjos, O., Santos, A. J. A., Estevinho, L. M., & Caldeira, I. (2016). FTIR-ATR spectroscopy applied to quality control of grape-derived spirits. *Food Chemistry*. https://doi.org/10.1016/j.foodchem.2016.02.128.

Aszyk, J., Byliński, H., Namieśnik, J., & Kot-Wasik, A. (2018). Main strategies, analytical trends and challenges in LC-MS and ambient mass spectrometry–based metabolomics. *TrAC - Trends in Analytical Chemistry*. https://doi.org/10.1016/j.trac.2018.09.010.

Barnaba, F. E., Bellincontro, A., & Mencarelli, F. (2014). Portable NIR-AOTF spectroscopy combined with winery FTIR spectroscopy for an easy, rapid, in-field monitoring of Sangiovese grape quality. *Journal of the Science of Food and Agriculture*. https://doi.org/10.1002/jsfa.6391.

Basalekou, M., Pappas, C., Tarantilis, P. A., & Kallithraka, S. (2020). Wine authenticity and traceability with the use of FT-IR. *Beverages*. https://doi.org/10.3390/beverages6020030.

Beale, D. J., Pinu, F. R., Kouremenos, K. A., Poojary, M. M., Narayana, V. K., Boughton, B. A., Kanojia, K., Dayalan, S., Jones, O. A. H., & Dias, D. A. (2018). Review of recent developments in GC–MS approaches to metabolomics-based research. *Metabolomics*. https://doi.org/10.1007/s11306-018-1449-2.

Belmonte-Sánchez, J. R., Romero-González, R., Martínez Vidal, J. L., Arrebola, F. J., & Garrido Frenich, A. (2020). 1H NMR and multi-technique data fusion as metabolomic tool for the classification of golden rums by multivariate statistical analysis. *Food Chemistry*. https://doi.org/10.1016/j.foodchem.2020.126363.

Bo, Y., Feng, J., Xu, J. J., Huang, Y., Cai, H., Cui, X., Dong, J., Ding, S., & Chen, Z. (2019). High-resolution pure shift NMR spectroscopy offers better metabolite discrimination in food quality analysis. *Food Research International*. https://doi.org/10.1016/j.foodres.2019.108574.

Bokulich, N. A., Collins, T. S., Masarweh, C., Allen, G., Heymann, H., Ebeler, S. E., & Millsa, D. A. (2016). Associations among wine grape microbiome, metabolome, and fermentation behavior suggest microbial contribution to regional wine characteristics. *MBio*. https://doi.org/10.1128/mBio.00631-16.

Bordiga, M., Lorenzo, C., Pardo, F., Salinas, M. R., Travaglia, F., Arlorio, M., Coïsson, J. D., & Garde-Cerdán, T. (2016). Factors influencing the formation of histaminol, hydroxytyrosol, tyrosol, and tryptophol in wine: Temperature, alcoholic degree, and amino acids concentration. *Food Chemistry*. https://doi.org/10.1016/j.foodchem.2015.11.112.

Borràs, E., Ferré, J., Boqué, R., Mestres, M., Aceña, L., & Busto, O. (2015). Data fusion methodologies for food and beverage authentication and quality assessment – A review. *Analytica Chimica Acta*. https://doi.org/10.1016/j.aca.2015.04.042.

Boulet, J. C., Williams, P., & Doco, T. (2007). A Fourier transform infrared spectroscopy study of wine polysaccharides. *Carbohydrate Polymers*. https://doi.org/10.1016/j.carbpol.2006.09.003.

Bowen, B. P., & Northen, T. R. (2010). Dealing with the unknown: Metabolomics and metabolite atlases. *Journal of the American Society for Mass Spectrometry*. https://doi.org/10.1016/j.jasms.2010.04.003.

Braconi, D., Bernardini, G., Millucci, L., & Santucci, A. (2018). Foodomics for human health: current status and perspectives. *Expert Review of Proteomics*. https://doi.org/10.1080/14789450.2018.1421072.

Brennan, L. (2014). NMR-based metabolomics: From sample preparation to applications in nutrition research. *Progress in Nuclear Magnetic Resonance Spectroscopy*. https://doi.org/10.1016/j.pnmrs.2014.09.001.

Brunius, C., Shi, L., & Landberg, R. (2016). Large-scale untargeted LC-MS metabolomics data correction using between-batch feature alignment and cluster-based within-batch signal intensity drift correction. *Metabolomics*. https://doi.org/10.1007/s11306-016-1124-4.

Cai, L., Rice, S., Koziel, J. A., & Dharmadhikari, M. (2017). Development of an automated method for selected aromas of red wines from cold-hardy grapes using solid-phase microextraction and gas chromatography-mass spectrometry-olfactometry. *Separations*. https://doi.org/10.3390/separations4030024.

Cassino, C., Tsolakis, C., Bonello, F., Gianotti, V., & Osella, D. (2019). Wine evolution during bottle aging, studied by 1H NMR spectroscopy and multivariate statistical analysis. *Food Research International*. https://doi.org/10.1016/j.foodres.2018.08.075.

Chaleckis, R., Meister, I., Zhang, P., & Wheelock, C. E. (2019). Challenges, progress and promises of metabolite annotation for LC–MS-based metabolomics. *Current Opinion in Biotechnology*. https://doi.org/10.1016/j.copbio.2018.07.010.

Charapitsa, S., Sytova, S., Kavalenka, A., Sobolenko, L., Kostyuk, N., Egorov, V., Leschev, S., Vetokhin, S., & Zayats, N. (2021). The study of the matrix effect on the method of direct determination of volatile compounds in a wide range of alcoholic beverages. *Food Control*. https://doi.org/10.1016/j.foodcont.2020.107528.

Chay, C., Elegado, F. B., Dizon, E. I., Hurtada, W. A., Norng, C., & Raymundo, L. C. (2017). PEffects of rice variety and fermentation method on the physiochemical and sensory properties of rice wine. *International Food Research Journal.*.

Ciepielowski, G., Pacholczyk-Sienicka, B., Frączek, T., Klajman, K., Paneth, P., & Albrecht, Ł. (2019). Comparison of quantitative NMR and IRMS for the authentication of 'Polish Vodka'. *Journal of the Science of Food and Agriculture*. https://doi.org/10.1002/jsfa.9168.

Cifuentes, A. (2012). Food analysis: Present, future, and foodomics. *ISRN Analytical Chemistry*. https://doi.org/10.5402/2012/801607.

Cioch-Skoneczny, M., Satora, P., Skoneczny, S., & Klimczak, K. (2020). Physicochemical characterization of wines produced using indigenous yeasts from cold climate grapes. *European Food Research and Technology*. https://doi.org/10.1007/s00217-020-03618-5.

Collins, T. S., Zweigenbaum, J., & Ebeler, S. E. (2014). Profiling of nonvolatiles in whiskeys using ultra high pressure liquid chromatography quadrupole time-of-flight mass spectrometry (UHPLC-QTOF MS). *Food Chemistry*. https://doi.org/10.1016/j.foodchem.2014.04.095.

Crisostomo, S. (2018, December 9). FDA records 21 deaths from lambanog. The Philippine Star. https://www.philstar.com/nation/2018/12/09/1875371/fda-records-21-deaths-lambanog.

D'Atri, V., Causon, T., Hernandez-Alba, O., Mutabazi, A., Veuthey, J. L., Cianferani, S., & Guillarme, D. (2018). Adding a new separation dimension to MS and LC–MS: What is the utility of ion mobility spectrometry?. *Journal of Separation Science*. https://doi.org/10.1002/jssc.201700919.

Day, L., & McSweeney, P. L. H. B. T.-R. M. in F. S. (2016). Beverages. Elsevier. https://doi.org/10.1016/B978-0-08-100596-5.03409-0.

Desbrosses, G., Steinhauser, D., Kopka, J., & Udvardi, M. (2005). Metabolome analysis using GC-MS. In Lotus Japonicus Handbook. https://doi.org/10.1007/1-4020-3735-X_17.

Dettmer, K., Aronov, P. A., & Hammock, B. D. (2007). Mass spectrometry-based metabolomics. *Mass Spectrometry Reviews*. https://doi.org/10.1002/mas.20108.

Diamantidou, D., Zotou, A., & Theodoridis, G. (2018). Wine and grape marc spirits metabolomics. *Metabolomics: Official Journal of the Metabolomic Society*. https://doi.org/10.1007/s11306-018-1458-1.

Donato, P., Rigano, F., Cacciola, F., Schure, M., Farnetti, S., Russo, M., Dugo, P., & Mondello, L. (2016). Comprehensive two-dimensional liquid chromatography–tandem mass spectrometry for the simultaneous determination of wine polyphenols and target contaminants. *Journal of Chromatography A*. https://doi.org/10.1016/j.chroma.2016.06.042.

Dzhimak, S. S., Basov, A. A., Buzko, V. Y., Kopytov, G. F., Kashaev, D. V., Shashkov, D. I., Shlapakov, M. S., & Baryshev, M. G. (2017). Determination of 13C/12C isotope ratio in alcohols of different origin by 1H nuclei NMR-spectroscopy. *Russian Physics Journal, 59*(10), 1710–1714. https://doi.org/10.1007/s11182-017-0965-2.

Esteki, M., Shahsavari, Z., & Simal-Gandara, J. (2018). Use of spectroscopic methods in combination with linear discriminant analysis for authentication of food products. *Food Control*. https://doi.org/10.1016/j.foodcont.2018.03.031.

Fan, S., Zhong, Q., Fauhl-Hassek, C., Pfister, M. K. H., Horn, B., & Huang, Z. (2018). Classification of Chinese wine varieties using 1H NMR spectroscopy combined with multivariate statistical analysis. *Food Control*. https://doi.org/10.1016/j.foodcont.2017.11.002.

FDA. (2018a). Food Advisories - BFAD Health Advisory 99-916 – Lambanog. https://ww2.fda.gov.ph/index.php/advisories-2/food-2/38601-bfad-health-advisory-99-916.

FDA. (2018b). Press Statement Lambanog. https://ww2.fda.gov.ph/index.php/news-and-events/545137-press-statement-lambanog.

Fotakis, C., & Zervou, M. (2016). NMR metabolic fingerprinting and chemometrics driven authentication of Greek grape marc spirits. *Food Chemistry*. https://doi.org/10.1016/j.foodchem.2015.10.002.

García-Cañas, V., & Simó, C. B. T.-R. M. in F. S. (2019). Food metabolomics—An overview. Elsevier. https://doi.org/10.1016/B978-0-08-100596-5.22666-8.

Garcia-Hernandez, C., Salvo-Comino, C., Martin-Pedrosa, F., Garcia-Cabezon, C., & Rodriguez-Mendez, M. L. (2020). Analysis of red wines using an electronic tongue and infrared spectroscopy. Correlations with phenolic content and color parameters. LWT. https://doi.org/10.1016/j.lwt.2019.108785.

Geana, E. I., Popescu, R., Costinel, D., Dinca, O. R., Ionete, R. E., Stefanescu, I., Artem, V., & Bala, C. (2016). Classification of red wines using suitable markers coupled with multivariate statistic analysis. *Food Chemistry*. https://doi.org/10.1016/j.foodchem.2015.07.112.

Gliszczyńska-Świgło, A., & Chmielewski, J. (2017). Electronic nose as a tool for monitoring the authenticity of food. A review. *Food Analytical Methods*. https://doi.org/10.1007/s12161-016-0739-4.

González-Robles, I. W., & Cook, D. J. (2016). The impact of maturation on concentrations of key odour active compounds which determine the aroma of tequila. *Journal of the Institute of Brewing*. https://doi.org/10.1002/jib.333.

Gougeon, L., da Costa, G., Guyon, F., & Richard, T. (2019). 1H NMR metabolomics applied to Bordeaux red wines. *Food Chemistry*. https://doi.org/10.1016/j.foodchem.2019.125257.

Hevia, K., Castro, R., Natera, R., González-García, J. A., Barroso, C. G., & Durán-Guerrero, E. (2016). Optimization of head space sorptive extraction to determine volatile compounds from oak wood in fortified wines. *Chromatographia*. https://doi.org/10.1007/s10337-016-3088-y.

Hird, S. J., Lau, B. P. Y., Schuhmacher, R., & Krska, R. (2014). Liquid chromatography-mass spectrometry for the determination of chemical contaminants in food. *TrAC – Trends in Analytical Chemistry*. https://doi.org/10.1016/j.trac.2014.04.005.

Hirst, M. B., & Richter, C. L. (2016). Review of aroma formation through metabolic pathways of Saccharomyces cerevisiae in beverage fermentations. *American Journal of Enology and Viticulture*. https://doi.org/10.5344/ajev.2016.15098.

Hu, B., Cao, Y., Zhu, J., Xu, W., & Wu, W. (2019). Analysis of metabolites in chardonnay dry white wine with various inactive yeasts by 1H NMR spectroscopy combined with pattern recognition analysis. *AMB Express*. https://doi.org/10.1186/s13568-019-0861-y.

Hu, B., Gao, J., Xu, S., Zhu, J., Fan, X., & Zhou, X. (2020). Quality evaluation of different varieties of dry red wine based on nuclear magnetic resonance metabolomics. *Applied Biological Chemistry, 63*(1), 24. https://doi.org/10.1186/s13765-020-00509-x.

Hu, B., Yue, Y., Zhu, Y., Wen, W., Zhang, F., & Hardie, J. W. (2015). Proton nuclear magnetic resonance-spectroscopic discrimination of wines reflects genetic homology of several different grape (V. vinifera L.) cultivars. PLoS ONE. 10.1371/journal.pone.0142840.

Hu, B., Zhao, Q., Yue, Y., Zhu, J., Lu, G., Li, H., & Zhang, F. (2016). 1H Nuclear magnetic resonance-based metabolomic study for Cabernet Sauvignon wines in different vintages. https://doi.org/10.7287/peerj.preprints.2332.

Hughey, C. A., McMinn, C. M., & Phung, J. (2016). Beeromics: from quality control to identification of differentially expressed compounds in beer. *Metabolomics*. https://doi.org/10.1007/s11306-015-0885-5.

Ickes, C. M., & Cadwallader, K. R. (2017). Effects of ethanol on flavor perception in alcoholic beverages. *Chemosensory Perception*. https://doi.org/10.1007/s12078-017-9238-2.

Jernigan, D., & Ross, C. S. (2020). The alcohol marketing landscape: Alcohol industry size, structure, strategies, and public health responses. *Journal of Studies on Alcohol and Drugs., Supplement*.

Johanningsmeier, S. D., Harris, G. K., & Klevorn, C. M. (2016). Metabolomic technologies for improving the quality of food: Practice and promise. *Annual Review of Food Science and Technology*. https://doi.org/10.1146/annurev-food-022814-015721.

Kamiloglu, S. (2019). Authenticity and traceability in beverages. *Food Chemistry*. https://doi.org/10.1016/j.foodchem.2018.10.091.

Kang, H.-R., Hwang, H.-J., Lee, J. E., & Kim, H. R. (2016). Quantitative analysis of volatile flavor components in Korean alcoholic beverage and Japanese sake using SPME-GC/MS. *Food Science and Biotechnology, 25*(4), 979–985. https://doi.org/10.1007/s10068-016-0159-7.

Karamanou, D. A., & Aliferis, K. A. (2020). Yeast (Saccharomyces cerevisiae) GC/EI/MS metabolomics dataset. *Data in Brief*. https://doi.org/10.1016/j.dib.2020.105208.

Kew, W., Goodall, I., & Uhrín, D. (2019). Analysis of Scotch Whisky by 1H NMR and chemometrics yields insight into its complex chemistry. *Food Chemistry*. https://doi.org/10.1016/j.foodchem.2019.125052.

Korban, A., Charapitsa, S., Čabala, R., Sobolenko, L., Egorov, V., & Sytova, S. (2021). Advanced GC–MS method for quality and safety control of alcoholic products. *Food Chemistry, 338*. https://doi.org/10.1016/j.foodchem.2020.128107.

Kuballa, T., Brunner, T. S., Thongpanchang, T., Walch, S. G., & Lachenmeier, D. W. (2018). Application of NMR for authentication of honey, beer and spices. *Current Opinion in Food Science*. https://doi.org/10.1016/j.cofs.2018.01.007.

Kueger, S., Steinhauser, D., Willmitzer, L., & Giavalisco, P. (2012). High-resolution plant metabolomics: From mass spectral features to metabolites and from whole-cell analysis to subcellular metabolite distributions. *Plant Journal*. https://doi.org/10.1111/j.1365-313X.2012.04902.x.

Kumar, K., Giehl, A., & Patz, C. D. (2019). Chemometric assisted Fourier Transform Infrared (FTIR) Spectroscopic analysis of fruit wine samples: Optimizing the initialization and convergence criteria in the non-negative factor analysis algorithm for developing a robust classification model. Spectrochimica Acta – Part A: Molecular and Biomolecular Spectroscopy. https://doi.org/10.1016/j.saa.2018.10.024.

Lachenmeier, D. W. (2016a). Advances in the detection of the adulteration of alcoholic beverages including unrecorded alcohol. *Advances in Food Authenticity Testing*. https://doi.org/10.1016/B978-0-08-100220-9.00021-7.

Lachenmeier, D. W. (2016b). Advances in the Detection of the Adulteration of Alcoholic Beverages Including Unrecorded Alcohol. *Advances in Food Authenticity Testing*. https://doi.org/10.1016/b978-0-08-100220-9.00021-7.

Lachenmeier, Dirk W., Richling, E., López, M. G., Frank, W., & Schreier, P. (2005). Multivariate analysis of FTIR and ion chromatographic data for the quality control of tequila. *Journal of Agricultural and Food Chemistry*. https://doi.org/10.1021/jf048637f.

Legras, J. L., Moreno-Garcia, J., Zara, S., Zara, G., Garcia-Martinez, T., Mauricio, J. C., Mannazzu, I., Coi, A. L., Zeidan, M. B., Dequin, S., Moreno, J., & Budroni, M. (2016). Flor yeast: New perspectives beyond wine aging. *Frontiers in Microbiology*. https://doi.org/10.3389/fmicb.2016.00503.

Lerma-García, M. J., Cortés, V., Talens, P., & Barat, J. M. (2018). Variety discrimination of fruits, edible plants, and other foodstuffs and beverages by infrared spectroscopy. *Comprehensive Analytical Chemistry*. https://doi.org/10.1016/bs.coac.2018.03.004.

Li, R., & Sun, Y. (2019). Effects of honey variety and non-saccharomyces cerevisiae on the flavor volatiles of mead. *Journal of the American Society of Brewing Chemists*. https://doi.org/10.1080/03610470.2018.1546072.

Li, R. Y., Zheng, X. W., Zhang, X., Yan, Z., Wang, X. Y., & Han, B. Z. (2018). Characterization of bacteria and yeasts isolated from traditional fermentation starter (Fen-Daqu) through a 1H NMR-based metabolomics approach. *Food Microbiology*. https://doi.org/10.1016/j.fm.2018.03.015.

Li, S., An, Y., Fu, W., Sun, X., Li, W., & Li, T. (2017). Changes in anthocyanins and volatile components of purple sweet potato fermented alcoholic beverage during aging. *Food Research International*. https://doi.org/10.1016/j.foodres.2017.08.041.

Li, Z., Wang, P. P., Huang, C. C., Shang, H., Pan, S. Y., & Li, X. J. (2014). Application of Vis/NIR spectroscopy for chinese liquor discrimination. *Food Analytical Methods*. https://doi.org/10.1007/s12161-013-9755-9.

Lim, M. (2019). Toxic coconut wine kills at least 11 people during Christmas celebrations in the Philippines. CNN. https://edition.cnn.com/2019/12/24/asia/lambanog-poisoning-philippines-intl-hnk-scli/index.html.

López-Ruiz, R., Romero-González, R., & Garrido Frenich, A. (2019). Metabolomics approaches for the determination of multiple contaminants in food. *Current Opinion in Food Science*. https://doi.org/10.1016/j.cofs.2019.08.006.

Lu, S. C., Liao, W. R., & Chen, S. F. (2018). Quantification of Trans-resveratrol in red wines using QuEChERS extraction combined with liquid chromatography-tandem mass spectrometry. *Analytical Sciences*. https://doi.org/10.2116/analsci.17P528.

Luo, Y., Kong, L., Xue, R., Wang, W., & Xia, X. (2020). Bitterness in alcoholic beverages: The profiles of perception, constituents, and contributors. *Trends in Food Science and Technology*. https://doi.org/10.1016/j.tifs.2019.12.026.

Magdas, D. A., Pirnau, A., Feher, I., Guyon, F., & Cozar, B. I. (2019). Alternative approach of applying 1H NMR in conjunction with chemometrics for wine classification. *LWT*. https://doi.org/10.1016/j.lwt.2019.04.054.

Mannu, A., Karabagias, I. K., Di Pietro, M. E., Baldino, S., Karabagias, V. K., & Badeka, A. V. (2020). 13C NMR-based chemical fingerprint for the varietal and geographical discrimination of wines. *Foods*. https://doi.org/10.3390/foods9081040.

Markley, John L, et al (2017). The Future of NMR-Based Metabolomics. *Current Opinion in Biotechnology, 43*, 34–40. https://doi.org/10.1016/j.copbio.2016.08.001.

Matyushin, D. D., Sholokhova, A. Y., Karnaeva, A. E., & Buryak, A. K. (2020). Various aspects of retention index usage for GC-MS library search: A statistical investigation using a diverse data set. *Chemometrics and Intelligent Laboratory Systems*. https://doi.org/10.1016/j.chemolab.2020.104042.

Medina, S., Perestrelo, R., Silva, P., Pereira, J. A. M., & Câmara, J. S. (2019). Current trends and recent advances on food authenticity technologies and chemometric approaches. *Trends in Food Science and Technology*. https://doi.org/10.1016/j.tifs.2019.01.017.

Miller, G. H., & Miller, G. H. (2019). Maturation. *Whisky Science*. https://doi.org/10.1007/978-3-030-13732-8_8.

Miyagawa, H., & Bamba, T. (2019). Comparison of sequential derivatization with concurrent methods for GC/MS-based metabolomics. *Journal of Bioscience and Bioengineering, 127*(2), 160–168. https://doi.org/10.1016/j.jbiosc.2018.07.015.

Monakhova, Y. B., Kuballa, T., & Lachenmeier, D. W. (2012). Nontargeted NMR analysis to rapidly detect hazardous substances in alcoholic beverages. *Applied Magnetic Resonance*. https://doi.org/10.1007/s00723-011-0309-2.

Morishima, K., Nakamura, N., Matsui, K., Tanaka, Y., Masunaga, H., Mori, S., Iwashita, T., li, X., & Shibayama, M. (2019). Formation of clusters in whiskies during the maturation process. Journal of Food Science. https://doi.org/10.1111/1750-3841.14398

Moros, G., Chatziioannou, A. C., Gika, H. G., Raikos, N., & Theodoridis, G. (2017). Investigation of the derivatization conditions for GC-MS metabolomics of biological samples. *Bioanalysis*. https://doi.org/10.4155/bio-2016-0224.

Moser, E., Laistler, E., Schmitt, F., & Kontaxis, G. (2017). Ultra-high field NMR and MRI-the role of magnet technology to increase sensitivity and specificity. *Frontiers in Physics*. https://doi.org/10.3389/fphy.2017.00033.

Neufeld, M., & Rehm, J. (2018). Effectiveness of policy changes to reduce harm from unrecorded alcohol in Russia between 2005 and now. *International Journal of Drug Policy*. https://doi.org/10.1016/j.drugpo.2017.09.006.

Newman, I. M., Qian, L., Tamrakar, N., & Zhang, B. B. (2018). Chemical composition and safety of unrecorded grain alcohol (Bai Jiu) samples from three provinces in China. *International Journal of Environmental Research and Public Health*. https://doi.org/10.3390/ijerph15122710.

Nichols, C. M., Dodds, J. N., Rose, B. S., Picache, J. A., Morris, C. B., Codreanu, S. G., May, J. C., Sherrod, S. D., & McLean, J. A. (2018). Untargeted molecular discovery in primary metabolism: Collision cross section as a molecular descriptor in ion mobility-mass spectrometry. *Analytical Chemistry*. https://doi.org/10.1021/acs.analchem.8b04322.

Nikolantonaki, M., Julien, P., Coelho, C., Roullier-Gall, C., Ballester, J., Schmitt-Kopplin, P., & Gougeon, R. D. (2018). Impact of glutathione on wines oxidative stability: A combined sensory and metabolomic study. *Frontiers in Chemistry*. https://doi.org/10.3389/fchem.2018.00182.

O'Sullivan, M. G. (2017). Chapter 13 - Sensory Properties of Beverage Products (Alcoholic and Nonalcoholic). In M. G. B. T.-A. H. for S. and C.-D. N. P. D. O'Sullivan (Ed.), Woodhead Publishing Series in Food Science, Technology and Nutrition (pp. 281–304). Woodhead Publishing. https://doi.org/10.1016/B978-0-08-100352-7.00013-0.

Palczak, J., Blumenthal, D., Rogeaux, M., & Delarue, J. (2019). Sensory complexity and its influence on hedonic responses: A systematic review of applications in food and beverages. *Food Quality and Preference*. https://doi.org/10.1016/j.foodqual.2018.06.002.

Papageorgiou, M., Lambropoulou, D., Morrison, C., Namieśnik, J., & Płotka-Wasylka, J. (2018). Direct solid phase microextraction combined with gas chromatography – Mass spectrometry for the determination of biogenic amines in wine. *Talanta*. https://doi.org/10.1016/j.talanta.2018.02.006.

Patra, S., Choudhary, R., Madhuri, R., & Sharma, P. K. (2019). Quality control of beverages for health safety: Starting from laboratory to the point-of-care detection techniques. In *Quality Control in the Beverage Industry: Volume 17: The Science of Beverages*. https://doi.org/10.1016/B978-0-12-816681-9.00002-3.

Pereira, L., Gomes, S., Barrias, S., Gomes, E., Baleiras-Couto, M., Fernandes, J., & Martins-Lopes, P. (2018). From the field to the bottle—An integrated strategy for wine authenticity. *Beverages*. https://doi.org/10.3390/beverages4040071.

Peterson, A. L., & Waterhouse, A. L. (2016). 1H NMR: A novel approach to determining the thermodynamic properties of acetaldehyde condensation reactions with glycerol, (+)-catechin, and glutathione in model wine. *Journal of Agricultural and Food Chemistry*. https://doi.org/10.1021/acs.jafc.6b02077.

Pinu, F. R., Villas-Boas, S. G., & Aggio, R. (2017). Analysis of intracellular metabolites from microorganisms: Quenching and extraction protocols. *Metabolites*. https://doi.org/10.3390/metabo7040053.

Polak, J., & Bartoszek, M. (2015). The study of antioxidant capacity of varieties of nalewka, a traditional Polish fruit liqueur, using EPR, NMR and UV-vis spectroscopy. *Journal of Food Composition and Analysis.*. https://doi.org/10.1016/j.jfca.2015.01.006.

Puxeu, M., Andorra, I., De Lamo-Castellví, S., & Ferrer-Gallego, R. (2019). Determination of nutrient supplementation by means of ATR-FTIR spectroscopy during wine fermentation. *Fermentation*. https://doi.org/10.3390/fermentation5030058.

Redruello, B., Ladero, V., del Rio, B., Fernández, M., Martin, M. C., & Alvarez, M. A. (2017). A UHPLC method for the simultaneous analysis of biogenic amines, amino acids and ammonium ions in beer. *Food Chemistry*. https://doi.org/10.1016/j.foodchem.2016.08.040.

Riu-Aumatell, M., Miró, P., Serra-Cayuela, A., Buxaderas, S., & López-Tamames, E. (2014). Assessment of the aroma profiles of low-alcohol beers using HS-SPME-GC-MS. *Food Research International*. https://doi.org/10.1016/j.foodres.2014.01.016.

Rizo, J., Guillén, D., Farrés, A., Díaz-Ruiz, G., Sánchez, S., Wacher, C., & Rodríguez-Sanoja, R. (2018). Omics in traditional vegetable fermented foods and beverages. *Critical Reviews in Food Science and Nutrition*. https://doi.org/10.1080/10408398.2018.1551189.

Rocha, W. F. de C., Sheen, D. A., & Bearden, D. W. (2018). Classification of samples from NMR-based metabolomics using principal components analysis and partial least squares with uncertainty estimation. Analytical and Bioanalytical Chemistry, 410 (24), 6305–6319. https://doi.org/10.1007/s00216-018-1240-2.

Rodgers, M. (2020). Chapter 15 - Food authenticity in the wine industry (D. B. T.-B. the F. of F. S. T. Detwiler (Ed.); pp. 147–157). Academic Press. https://doi.org/https://doi.org/10.1016/B978-0-12-818956-6.00015-4.

Rugani, B. (2018). Environmental externalities in global trade for wine and other alcoholic beverages. *Encyclopedia of Food Security and Sustainability*. https://doi.org/10.1016/B978-0-08-100596-5.21974-4.

Sagandykova, G. N., Alimzhanova, M. B., Nurzhanova, Y. T., & Kenessov, B. (2017). Determination of semi-volatile additives in wines using SPME and GC–MS. *Food Chemistry*. https://doi.org/10.1016/j.foodchem.2016.09.164.

Sánchez-Estébanez, C., Ferrero, S., Alvarez, C. M., Villafañe, F., Caballero, I., & Blanco, C. A. (2018). Nuclear magnetic resonance methodology for the analysis of regular and non-alcoholic lager beers. *Food Analytical Methods*. https://doi.org/10.1007/s12161-017-0953-8.

Santos, M. C., Nunes, C., Ferreira, A. S., Jourdes, M., Teissedre, P. L., Rodrigues, A., Amado, O., Saraiva, J. A., & Coimbra, M. A. (2019). Comparison of high pressure treatment with conventional red wine aging processes: impact on phenolic composition. *Food Research International*. https://doi.org/10.1016/j.foodres.2018.08.018.

Singh, D., Lee, S., & Lee, C. H. (2017). Metabolomics for empirical delineation of the traditional Korean fermented foods and beverages. *Trends in Food Science and Technology*. https://doi.org/10.1016/j.tifs.2017.01.001.

Spinelli, F. R., Cargnel, G. J., Drehmer, A. P., Blatt, C., Baptistão, M., & Vanderlinde, R. (2019). Analysis of allergenic residues in wines by triple quadrupole LCMS. *BIO Web of Conferences*. https://doi.org/10.1051/bioconf/20191204012.

Spraul, M., Link, M., Schaefer, H., Fang, F., & Schuetz, B. (2015). Wine analysis to check quality and authenticity by fully-automated 1 H-NMR. *BIO Web of Conferences*. https://doi.org/10.1051/bioconf/20150502022.

Spyros, A. (2016). Application of NMR in food analysis. *Nuclear Magnetic Resonance*. https://doi.org/10.1039/9781782624103-00269.

Stelick, A., & Dando, R. (2018). Thinking outside the booth — the eating environment, context and ecological validity in sensory and consumer research. *Current Opinion in Food Science*. https://doi.org/10.1016/j.cofs.2018.05.005.

Stupak, M., Kocourek, V., Kolouchova, I., & Hajslova, J. (2017). Rapid approach for the determination of alcoholic strength and overall quality check of various spirit drinks and wines using GC–MS. *Food Control*. https://doi.org/10.1016/j.foodcont.2017.05.008.

Tada, I., Tsugawa, H., Meister, I., Zhang, P., Shu, R., Katsumi, R., Wheelock, C. E., Arita, M., & Chaleckis, R. (2019). Creating a reliable mass spectral–retention time library for all ion fragmentation-based metabolomics. *Metabolites*. https://doi.org/10.3390/metabo9110251.

Takase, H., Sasaki, K., Shinmori, H., Shinohara, A., Mochizuki, C., Kobayashi, H., Saito, H., Matsuo, H., Suzuki, S., & Takata, R. (2015). Analysis of rotundone in japanese syrah grapes and wines using stir bar sorptive extraction (SBSE) with heart-cutting two-dimensional GC-MS. *American Journal of Enology and Viticulture*. https://doi.org/10.5344/ajev.2015.14118.

Taniguchi, M., Shimotori, A., & Fukusaki, E. (2019). Enantioselective amino acid profile improves metabolomics-based sensory prediction of Japanese sake. *Food Science and Technology Research*. https://doi.org/10.3136/fstr.25.775.

Teixeira dos Santos, C. A., Páscoa, R. N. M. J., Porto, P. A. L. S., Cerdeira, A. L., & Lopes, J. A. (2016). Application of Fourier-transform infrared spectroscopy for the determination of chloride and sulfate in wines. *LWT – Food Science and Technology*. https://doi.org/10.1016/j.lwt.2015.11.050.

Trimigno, A., Marincola, F. C., Dellarosa, N., Picone, G., & Laghi, L. (2015). Definition of food quality by NMR-based foodomics. *Current Opinion in Food Science*. https://doi.org/10.1016/j.cofs.2015.06.008.

Uzhel, A. S., Borodina, A. N., Gorbovskaya, A. V., Shpigun, O. A., & Zatirakha, A. V. (2020). Determination of full organic acid profiles in fruit juices and alcoholic beverages using novel chemically derivatized hyperbranched anion exchanger. *Journal of Food Composition and Analysis*. 103674.

Versari, A., Parpinello, G. P., Scazzina, F., & Del Rio, D. (2010). Prediction of total antioxidant capacity of red wine by Fourier transform infrared spectroscopy. *Food Control*. https://doi.org/10.1016/j.foodcont.2009.11.001.

Vinaixa, M., Schymanski, E. L., Neumann, S., Navarro, M., Salek, R. M., & Yanes, O. (2016). Mass spectral databases for LC/MS- and GC/MS-based metabolomics: State of the field and future prospects. *TrAC – Trends in Analytical Chemistry*. https://doi.org/10.1016/j.trac.2015.09.005.

Walker, G., & Stewart, G. (2016). Saccharomyces cerevisiae in the Production of Fermented Beverages. *Beverages*. https://doi.org/10.3390/beverages2040030.

Wang, F., Shao, C., Chen, Q., Meng, T., & Li, C. (2019). Application on sensory prediction of Chinese Moutai-flavour liquor based on ATR-FTIR. E3S Web of Conferences. https://doi.org/10.1051/e3sconf/20197903001.

Wang, L., Sun, D. W., Pu, H., & Cheng, J. H. (2017). Quality analysis, classification, and authentication of liquid foods by near-infrared spectroscopy: A review of recent research developments. *Critical Reviews in Food Science and Nutrition*. https://doi.org/10.1080/10408398.2015.1115954.

Wardencki, W. (2018). Alcoholic Beverages. In Reference Module in Chemistry, Molecular Sciences and Chemical Engineering. Elsevier. https://doi.org/10.1016/B978-0-12-409547-2.14330-6.

Wishart, D. S. (2019). NMR metabolomics: A look ahead. *Journal of Magnetic Resonance*. https://doi.org/10.1016/j.jmr.2019.07.013.

Wiwanitkit, V. (2019). Alcoholic beverage production in Indochina: Local wisdom, safety, quality, and legal control. *Production and Management of Beverages*. https://doi.org/10.1016/b978-0-12-815260-7.00013-4.

Yadav, P. K., & Sharma, R. M. (2019). Classification of illicit liquors based on their geographic origin using Attenuated total reflectance (ATR) – Fourier transform infrared (FT-IR) spectroscopy and chemometrics. *Forensic Science International*. https://doi.org/10.1016/j.forsciint.2018.12.017.

Yıldırım, H. K. (2021). 2 - Insights into the role of yeasts in alcoholic beverages. In R. C. B. T.-M. B. in F. and H. Ray (Ed.), Applied biotechnology reviews (pp. 21–52). Academic Press. https://doi.org/10.1016/B978-0-12-819813-1.00002-5.

Zeki, Ö. C., Eylem, C. C., Reçber, T., Kır, S., & Nemutlu, E. (2020). Integration of GC–MS and LC–MS for untargeted metabolomics profiling. *Journal of Pharmaceutical and Biomedical Analysis*. https://doi.org/10.1016/j.jpba.2020.113509.

Zerbib, M., Mazauric, J. P., Meudec, E., Le Guernevé, C., Lepak, A., Nidetzky, B., Cheynier, V., Terrier, N., & Saucier, C. (2018). New flavanol O-glycosides in grape and wine. *Food Chemistry*. https://doi.org/10.1016/j.foodchem.2018.06.019.

Zhang, J., Hu, Q., Yu, Q., Chen, Y., Zhao, Y., & Qie, M. B. T.-R. M. in F. S. (2020). Metabolomics Analysis in Food Authentication. Elsevier. https://doi.org/https://doi.org/10.1016/B978-0-08-100596-5.22793-5.

Zhang, W., & Xue, J. (2016). Economically motivated food fraud and adulteration in China: An analysis based on 1553 media reports. *Food Control*. https://doi.org/10.1016/j.foodcont.2016.03.004.

Zhou, Q., Qian, Y., & Qian, M. C. (2015). Analysis of volatile phenols in alcoholic beverage by ethylene glycol-polydimethylsiloxane based stir bar sorptive extraction and gas chromatography-mass spectrometry. *Journal of Chromatography A*. https://doi.org/10.1016/j.chroma.2015.02.064.

Zhu, J., Hu, B., Lu, J., & Xu, S. (2018). Analysis of metabolites in cabernet sauvignon and shiraz dry red wines from Shanxi by 1H NMR spectroscopy combined with pattern recognition analysis. *Open Chemistry*. https://doi.org/10.1515/chem-2018-0052.