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Research Article

Correlation Between Colorimetric Properties and Phenolic Compounds in Aged Vinegars

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ABSTRACT

The aged vinegars studied, which are *Sherry* vinegars from Jerez, Spain, with varying aging periods, are protected under the Denomination of Origin (DO). The objectives of this study were to (i) examine the impact of aging on the polyphenolic compound content and color properties of *Sherry* vinegars, and (ii) determine the correlation between phenolic compounds and color properties in these samples. Phenolic compounds were identified and quantified using LC-MS/MS technique, and correlation analysis was conducted between phenolic content and color properties. The results showed a statistically significant decrease in total phenolic content with increased aging. Additionally, all colorimetric properties exhibited significant changes across the samples. A notable shift from yellow to red hues was observed in the vinegars. Significant reductions in caffeic acid and gallic acid content were also found due to aging. This study is the first to analyze the correlation between phenolics and color properties in *Sherry* vinegars, revealing the influence of phenolic compounds on their color characteristics.

Keywords: Sherry vinegar, phenolics, LC-MS/MS, color properties

Yıllandırılmış Sirkelerde Kolorimetrik Özellikler ile Fenolik Bileşikler Arasındaki Korelasyon

ÖZ

Araştırma kapsamında incelenen örnekler, İspanya'nın Jerez bölgesinden farklı yıllandırma sürelerine sahip Menşei Sistemi altında korunan *Sherry* sirkeleridir. Bu çalışmanın amaçları (i) yıllandırmanın *Sherry* sirkelerinin polifenolik bileşik içeriği ve renk özellikleri üzerindeki etkisini incelemek ve (ii) bu örneklerdeki fenolik bileşikler ile renk özellikleri arasındaki korelasyonu belirlemektir. Fenolik bileşikler, LC-MS/MS tekniği kullanılarak tanımlandı ve fenolik içerik ile renk özellikleri arasında korelasyon analizi yapılmıştır. Sonuçlar yıllandırmanın artmasıyla birlikte analiz edilen fenolik içeriğin toplamında istatistiksel olarak anlamlı bir azalma olduğunu göstermiştir. Ayrıca numuneler arasında tüm kolorimetrik özelliklerde önemli değişiklikler gözlenmiştir. Sirke örneklerinde sarı tonlardan kırmızı tonlara gözle görülür bir geçiş gözlenmiştir. Yıllanmaya bağlı olarak kafeik asit ve gallik asit içeriğinde de önemli azalmalar bulunmuştur. Bu çalışma, *Sherry* sirkelerindeki fenolikler ile renk özellikleri arasındaki ilişkiyi analiz etmiş olup ve fenolik bileşiklerin renk özellikleri üzerindeki etkisini ortaya koyan ilk çalışma olmuştur.

Anahtar kelime: Sherry sirkesi, fenolikler, LC-MS/MS, renk özellikleri

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Introduction

Vinegars are one of the important seasoning foods consumed for a long time throughout history (Liang et al. 2016). Vinegars can be primarily categorized into two main types based on differences in raw materials and processing techniques: grain vinegars and fruit vinegars (Xia et al. 2018). Sherry vinegar, Spain's most popular and valuable fruit vinegar, is also one of the most famous vinegars worldwide. It stands as a highly esteemed and top-tier product hailing from the Denomination of Origin (DO) region in the southwestern part of Spain. This prestigious designation, protected by both Spain and the European Union (Tesfaye et al. 2002a). Sherry vinegars crafted within geographically indicated regions, is derived from Sherry wines meticulously produced using the traditional aging method known as "Soleras and Criaderas" (Andalucia and Deporte 2006). This time-honored technique lends a distinctive character and quality to the vinegar, reflecting the rich heritage of the region.

One of the features that make vinegar a unique product is its chemical composition. Phenolic compounds are one of the significant components of vinegar which mostly affect the organoleptic properties and quality. These compounds in vinegar are initially sourced from raw material (grapes) and subsequently wine during acidification process (Cosme et al. 2018). In the fermentation and aging periods, phenolic compounds can undergo some reactions with other compounds of wine such as tannins. These reactions are playing an important role in defining the characteristic properties of vinegars. Color, appearance, flavor and also taste can be changed according to the change in the content and variability of phenolic compounds (Uysal et al. 2023). In addition to phenolic compounds, color properties of vinegar depend on the origin of grape varieties, then fermentation process and type of aging process. Thus, the aging process can be a determinative feature on *Sherry* vinegar because of time and unique process. "*Solera and Criadera*" is a type of production process which is carried out according to a dynamic aging system from the South region of Spain (Valcárcel-Muñoz et al. 2023). Therefore, knowing the chemical process that occurs on the phenolic components during the production processes of special vinegars is one of the areas that need to be studied in order to reveal the effect of aging and acetification processes and to show how it changes their organoleptic properties.

There have been some studies on the effect of acetification process on phenolics (Andlauer et al. 2000), evaluation of bioactive compounds of Shanxi aged vinegars (Xia et al. 2018), changes in antioxidant components during aging of traditional balsamic vinegar (Verzelloni et al. 2010), correlation of antioxidant and color profile of balsamic vinegars (Sinanoglou et al. 2018), antioxidant characteristics of traditional Zhenjiang vinegars during aging (Zhao et al. 2018), changes in color and phenolic compounds during aging of *Sherry* white wine (Ortega et al. 2003), evaluation of phenolics during experimental aging within 180 days of Sherry vinegars (Tesfaye et al. 2002b). As a result of the examination of the studies conducted and to our best knowledge, no study has been found examining the relationship between the phenolic component and organoleptic properties of *Sherry* vinegars, which change with long-term aging period in a wood cask.

The aim of this study is to show how aging affects the relationship between color properties and phenolic component composition of *Sherry* vinegars of different ages. Secondly, this analytical approach aimed to elucidate any potential connections between the color characteristics of the aged vinegars and the concentrations of some phenolic compounds, providing deeper insights into the compositional attributes of the samples.

Materials and Method

Reagents and chemicals

Reagents and chemicals for chromatographic analysis were purchased from Panreac Applichem (Darmstadt, Germany). Myricetin-3-*O*-glucoside, quercetin-3-*O*-glucoside; quercetin-3-*O*-galactoside; quercetin-3-*O*-rutinoside; quercetin-3-*O*-glucuronide; quercetin-3-*O*-glucopyranoside; luteolin-7-*O*-glucoside; caffeic acid, chlorogenic acid, and gallic acid were purchased from Cymit Química (Barcelona, Spain).

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Samples

Three conventional sherry vinegar samples aged at different years (*Crianza*, *Reserva*, and *Gran Reserva*) were supplied from Sur de Espana (Jerez de la Frontera, Cádiz, Spain). All the samples were supplied in duplicate (from two batches of the same kind of vinegar). The entirety of the sherry vinegar specimens was exclusively crafted utilizing Sherry Wine's provenance within the Protected Designation of Origin (PDO) framework, which entail distinct aging durations as stipulated by the PDO guidelines. The maturation process of sherry vinegar transpires within wooden casks, following the traditional "*Criadera y Solera*" system. Specifically, the examined samples underwent aging within American oak barrels, employing the "Criadera y Solera" methodology. As per the specifications outlined by the PDO for sherry wine vinegars, the aging process must adhere to the following guidelines: (i) For *Crianza* vinegars, the aging duration must be at least of 6 months; (ii) Regarding Reserva sherry vinegar, the aging period must maintain a minimum of 2 years; (iii) Gran Reserva sherry vinegar necessitates an aging period surpassing 10 years. In this study, the aging period of analyzed samples were approximately as follows: Crianza (8 months), Reserva (3 years), and Gran reserva (12 years).

Color properties

The chromatic attributes, encompassing tonality, color intensity, and color density, within the sherry vinegar specimens were evaluated in accordance with the methodology delineated by the International Organization of Vine and Wine (OIV 2022). Absorbance measurements were conducted utilizing a UV/VIS spectrophotometer (PG Instruments T80, UK). The colorimetric features of the vinegar samples were assessed at specific wavelengths, namely 420 nm (representing yellow components), 520 nm (depicting red components), and 620 nm (indicating blue components). Subsequently, calculations for color intensity (CI), tonality (T), and color density (D) were executed utilizing the ensuing formulas:

$$CI = A420 + A520 + A620$$

$$T = A420/A520$$

$$D = A420 + A520$$

The Glories color index percentages corresponding to yellow (Y%), red (R%), and blue (B%) hues present in the vinegars were computed utilizing the spectral data acquired at wavelengths of 420 nm, 520 nm, and 620 nm, respectively (Glories 1984).

Phenolic compounds

Polyphenol composition of the samples was conducted utilizing a Shimadzu LC-MS/MS 8050 triple quadrupole mass spectrometer outfitted with an electrospray ionization (ESI) source. The vinegar samples were filtered through a 0,45 µm pore size membrane filter before injection. The mobile phase utilized for the separation of polyphenols comprised two solvent systems: (i) Solvent A, consisting of water/formic acid (99,9:0,1, v/v), and (ii) Solvent B, comprising acetonitrile/formic acid (99,9:0,1, v/v). A sample volume of 10 µL was injected for analysis. The characterization of individual components was conducted by assessing their retention times and precise molecular masses. Furthermore, concentrations of each compound were determined through external calibration using corresponding standard compounds. To achieve this, stock solutions of all individual standards were prepared with the extractant across a concentration range of 0.1, 0.3, 0.5, 0.8, and 1 mg L^{-1}. This calibration procedure facilitated the accurate quantification of the concentrations of the target compounds within the sample. The analysis was performed in duplicate.

Statistical analysis

Statistical analysis was carried out using XLSTAT Premium 2016 (Addinsoft, New York, NY). One-way analysis of variance (ANOVA) was employed to assess significant differences in colorimetric characteristics and polyphenol compounds among the vinegar samples.

Subsequently, Tukey's multiple range tests were applied as a post-hoc analysis, with significance typically inferred when the resulting *p*-value was below 0,05. Furthermore, a correlation analysis was undertaken to investigate potential relationships between the colorimetric parameters of vinegars and the levels of polyphenols. Correlation coefficients resulting from this analysis were reported to ascertain the degree of association between these variables.

Results and Discussion

Color properties

Color intensity (CI), tonality (T), color density (D), and percentages of yellow, red, and blue hues (%) in vinegar samples were measured using a spectrophotometer. The specific values for each colorimetric parameter of the samples are provided in Table 1. As can be seen from the table, the highest yellow characteristics and the lowest red tones of the samples were found for *V1* (*Crianza*, 8 months) sample with the percentages of 65.70% and 24.93%, respectively. The lowest yellow and the highest red color were also obtained for *V2* (*Reserva*, 3 years) sample with the amount of 60.50% and 28.57%.

On the other hand, the yellow and red components of the most aged *V3* (*Gran Reserva*, 12 years) sample were calculated as the percentage of 62.51 and 26.83, respectively. As can be seen from the table, an increase in the absorbance values of A420nm and A520nm was observed for the samples of V2 and V3 compared to V1. These changes observed in the yellow (decrease in the amount) and red pigments (increase in the percentage) during the aging process of samples (from V1 to V2 and V3) can be attributed to browning reactions such as Maillard. Especially the initial decrease in the sample of V1 to V2 may be released due to amino acids may involve in different reactions which result in the formation of Maillard compounds and the similar change in browning was observed in another research studied by (Palacios et al. 2002).

The formation of these browning compounds during the aging period resulted in color change leading to an increase in CI (4,68%) and D (4,17%) values for V2 sample. A similar change for CI (4,45%) and D (3,97%) values was obtained for V3 sample. On the other hand, although there was a statistically significant difference between the samples of V1-V2 (from 9,38% to 10,93) and V1-V3 (from 9,38% to 10,66%) in blue tones, not much change was observed in the percentage of the color component.

The significant color change in yellow and red components, and the slight change in blue component can be considered as a result proving that these non-enzymatic reactions occur during the aging process. The reactions described play a significant role in the notable increase in total absorbance (CI) observed during the physicochemical phase, as outlined in Table 1. These reactions contribute to the enhancement of the color properties. Similarly, browning in color and an increase in red tone were also observed in Shanxi vinegar, another example of traditional aged vinegar having geographical indication (PGI) certificate which is produced in Shanxi Province, China (Zhu et al. 2020). Most of the brown-colored compounds that can be formed by caramelization along with Maillard are melanoidins (Bozkurt et al. 1999).

The formation of the basic structure of melanoidins is related to the formation of 5-hydroxymethyl-2-furfural (HMF) and furfural during aging (Delgado-Andrade and Morales 2005, Tagliazucchi et al. 2010). In vinegar characterized by high sugar concentrations and low water content, the formation of HMF and furfurals occurs even under ambient conditions, including room temperature (Yang et al. 2014). A notable disparity in tonality values was observed between the V1-V2 and V1-V3 samples. This discrepancy can be attributed to an increase in absorbance values at 520 nm (A520nm) and intensification of red coloration due to browning compounds.

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Table 1. Colorimetric characteristics of wine-vinegar (V) samples

Vinegar Type	A_{420}¶	A_{520}¶	A_{620}¶	CI	Tonality	D	Y¶ (%)	R¶ (%)	B¶ (%)
ANOVA†	***	***	***	***	***	***	***	***	***
V1 (8 M‡)	2,053 ± 0,01 c	0,779 ± 0,02 c	0,293 ± 0,03 c	3,13 ± 0,02 c	2,64 ± 0,01 a	2,83 ± 0,01 c	65,70 ± 0,03 a	24,93 ± 0,05 c	9,38 ± 0,06 b
V2 (3 Y§)	2,829 ± 0,01 a	1,336 ± 0,01 a	0,511 ± 0,01 a	4,68 ± 0,01 a	2,12 ± 0,02 c	4,17 ± 0,02 a	60,50 ± 0,04 c	28,57 ± 0,06 a	10,93 ± 0,07 a
V3 (12 Y)	2,779 ± 0,02 b	1,193 ± 0,02 b	0,474 ± 0,01 b	4,45 ± 0,02 b	2,33 ± 0,01 b	3,97 ± 0,01 b	62,51 ± 0,06 b	26,83 ± 0,04 b	10,66 ± 0,05 a

*, **, ***, significant at $p < 0.05$, $p < 0.01$, and $p < 0.001$, respectively. Mean values obtained from the triplicate analysis of all vinegar samples. Values followed by the same letter, within the same row, were not significantly different ($p > 0.05$), according to Tukey's least significant difference test. CI: Color intensity, D: Color density, Y¶: yellow (%), R: red (%), B: blue (%), M‡: month, Y§: year.

Phenolic compounds

In addition to colorimetric properties, phenolic compounds in vinegar samples were analyzed using LC-MS/MS. The concentrations of these phenolic compounds in all vinegar samples are presented in Table 2. It is evident from the table that most of the phenolic compounds were initially found at high concentrations in the V1 sample, with subsequent decreases observed during the aging period. For instance, the concentration of Quercetin-O-3-glucoside was 45,21 µg kg^{-1} in the V1 sample but was not detected (ND) in the V3 sample. A significant reduction was also noted in the content of Quercetin-O-3-glucuronide from 1029,05 µg kg^{-1} in V1 to 61,51 µg kg^{-1} in V3. Furthermore, notable differences were observed between V1-V2 and V1-V3 samples for gallic acid and caffeic acid. The concentration of gallic acid decreased from 641,19 µg kg^{-1} in V1 to 219,97 µg kg^{-1} in V2 and 200,6 µg kg^{-1} in V3. Conversely, for caffeic acid, the concentrations were determined as 1049,8 µg kg^{-1}, 213,98 µg kg^{-1}, and 95,58 µg kg^{-1} for V1, V2, and V3 samples, respectively. These findings highlight dynamic changes in the phenolic composition of vinegar samples during the aging process, which may contribute to alterations in chemical profiles.

Statistically significant results were found for total phenolic compounds between the samples. While at least aged sample (V1) has the highest phenolic compound content (2896,06), the most aged sample (V3) has the less content of phenolic compounds (296,18). Moreover, there is a significant change between the samples of V1 and V2 with a content of 990,71 (V2). The total phenolic compounds result shows a high relation between aging period and the content of phenolics.

On the other hand, as Luteolin-7-O-glucoside was found in a very small amount but also decrease during aging period was observed from 5,26 (V1) to 1,72 (V3). Quercetin-3-O-rutinoside cannot be detected (ND) in all samples. While chlorogenic acid, one of the phenolic acids, was not seen (ND) in the V1 and V3 samples, it was seen in the amount of 115,85 µg kg^{-1} in the V2 sample. Eriotricin and hesperidin compounds from flavanones were also found in the amount of 88,16 and 64,63 µg kg^{-1}, respectively in only V2 sample.

In another study, a similar change and progressive loss in phenolic compounds, especially monomeric flavanols such as catechin, has been observed during traditional balsamic vinegar. It was also stated in the study that some reactions involving flavonoids and tannins may lead to subsequent polycondensation of other flavonoid molecules. As a result of these reactions, while a decrease was observed in monomeric flavonoids in aging of traditional balsamic vinegar, an increase was also detected in polymeric phenolic compounds.

Table 2. Mass spectral characteristics and concentration of phenolic compounds (μg Kg^{-1}) present in the vinegar samples.

Code	Compound	$[M-H]^-$ (m/z)	MS/MS (m/z)	ANOVA‡	V1	V2	V3
Flavones							
1	Luteolin-7-*O*-glucoside	448,90	†**287,10**/153,10	*	5,26 ± 0,01 a	2,72 ± 0,05 b	1,72 ± 0,01 b
Flavanones							
2	Eriotricin	595,20	**287,05**/150,09	***	‡ND b	88,16 ± 0,01 a	ND b
3	Hesperidin	609,20	**301**/163,90	***	ND b	64,63 ± 0,01 a	ND b
Flavonols							
4	Myricetin-3-*O*-glucoside	481,10	**319,10**/273,10	***	28,56 ± 0,02 b	45,09 ± 0,01 a	28,67 ± 0,01 b
5	Quercetin-3-*O*-glucoside	463,25	**300,15**/271.15	*	45,21 ± 0,03 a	26,12 ± 0,01 b	ND c
6	Quercetin-3-*O*-glucuronide	478,95	**303,05**/229	***	1029,05 ± 0,02 a	144,43 ± 0,01 b	61,51 ± 0,02 b
7	Quercetin-3-*O*-galactoside	465	**303,10**/229,1	***	61,90 ± 0,01 a	38,75 ± 0,02 b	ND c
8	Quercetin-3-*O*-rutinoside	608,90	301/**299,95**	NS	ND a	tr¶ a	ND a
9	Quercetin-3-*O*-glucopyranoside	463,10	300,95/**300**	***	35,09 ± 0,02 a	31,01 ± 0,02 a	ND b
Phenolic acids and derivatives							
10	Gallic acid	169,10	**124,95**/124,30	NS	641,19 ± 0,01 a	219,97 ± 0,03 b	200,6 ± 0,03 b
11	Chlorogenic acid	353,30	**191,05**/92,95	NS	ND b	115,85 ± 0,01 a	ND b
12	Caffeic acid	179,10	**135**/134/106.95	***	1049,8 ± 0,02 a	213,98 ± 0,01 b	95,58 ± 0,01 b
	Total of polyphenolic compounds			***	2896,06 ± 0,04 a	990,71 ± 0,02 b	296,18 ± 0,02 c

†The m/z values of the dominant ions are highlighted using bold font. ¶tr: trace. ‡ND: not detected. ǂNS: not significant at $p < 0,05$; *, **, ***, significant at $p < 0,05$, $p < 0,01$, and $p < 0,001$, respectively. Mean values obtained from the duplicate analysis of all vinegar samples. Values followed by the same letter, within the same row, were not significantly different ($p > 0,05$), according to Tukey's least significant difference test.

During aging of balsamic vinegar, the amount of total polyphenols decreased from 1500,6 mg/kg to 1321,4 mg/kg (Verzelloni et al. 2010). The formation of some of the phenolic compounds (such as Eriotricin, hesperidin or flavonols; myricetin-3-O-glucoside) in V2 samples (in three years) during aging may be explained by the extraction of phenolic compounds from the oak butt (Tesfaye et al. 2002a).

Correlation between color properties and phenolic compounds

Pearson's correlation analysis was conducted to examine the relationship between the color properties (such as color intensity, density, percentages of colors (yellow, red, and blue) and the phenolic compounds (Quercetin-O-3-glucuronide, Quercetin-O-3-galactoside, gallic acid, caffeic acid, and total phenolics) in all vinegar samples. Table 3 presents the correlation coefficient matrix of this analysis between the color parameters and phenolic compounds. A statistically significant ($p < 0,05$) positive relationship was observed, indicating high correlation coefficients between certain phenolic compounds such as Quercetin-3-O-glucuronide-caffeic acid (r=0,999); Quercetin-3-O-glucuronide-gallic acid (r=0,999); gallic acid-caffeic acid (r=0,997).

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Table 3. Pearson correlation coefficients matrix among the color properties and phenolic compounds determined in the samples

Variables	Y (%)	R (%)	B (%)	CI	D	Quercetin-3-O-glucuronide	Quercetin-3-O-galactoside	Gallic acid	Caffeic acid	Total phenolic
Y (%)	**1*****†	-0,995	-0,974	-0,968	-0,968	0,891	0,488	0,908	0,874	0,794
R (%)	-0,995	**1****	0,945	0,936	0,936	-0,839	-0,394	-0,859	-0,818	-0,725
B (%)	-0,974	0,945	**1****	**1,000***	**1,000***	-0,971	-0,674	-0,980	-0,962	-0,911
IC	-0,968	0,936	**1,000***	**1****	**1,000****	-0,977	-0,692	-0,984	-0,968	-0,922
D	-0,968	0,936	**1,000***	**1,000****	**1****	-0,977	-0,692	-0,984	-0,968	-0,921
Quercetin-3-O-glucuronide	0,891	-0,839	-0,971	-0,977	-0,977	**1****	0,831	**0,999***	**0,999***	0,983
Quercetin-3-O-galactoside	0,488	-0,394	-0,674	-0,692	-0,692	0,831	**1****	0,809	0,850	0,918
Gallic acid	0,908	-0,859	-0,980	-0,984	-0,984	**0,999***	0,809	**1****	**0,997***	0,975
Caffeic acid	0,874	-0,818	-0,962	-0,968	-0,968	**0,999***	0,850	**0,997***	**1****	0,989
Total phenolic	0,794	-0,725	-0,911	-0,922	-0,921	0,983	0,918	0,975	0,989	**1****

†Values in bold font are statistically significant **, **, and *** at p < 0,05, 0,01, and 0,001,* respectively.

The decrease in the amounts of these flavonol components and phenolic acid components in the vinegar samples over the aging period contributes to their correlation with each other. Although a decrease in the Quercetin-3-O-galactoside component is observed due to aging, a correlation with a high coefficient may not have been found due to its small amount. Based on the findings, it can be inferred that all free mono-phenolic compounds underwent a series of reactions, leading to their disappearance or presence in very low amounts in the vinegar sample.

A significant positive correlation was observed among some color properties of the vinegar samples. Specifically, a high positive correlation was noted between blue pigment–color intensity (r = 1,000) and blue pigment-density (r = 1,000). Similarly, the color intensity and density of the samples exhibited a highly significant and positive correlation (r = 1,000). These correlation findings align with the earlier observations regarding changes in blue (%), color intensity, and density properties of the vinegar samples as the aging period increases. Although a strong negative correlation (with *r*= -0,995) was observed in yellow and red colors, it was not found to be statistically significant.

No statistically significant relationship was detected between (i) color properties and (ii) phenolic compounds. This absence of correlation could potentially be attributed to little increase in yellow (%) pigment (from 60,50 to 62,51) and minor decline in red (%) pigment (from 28,57 to 26.83) between V2-V3 samples during aging. This observed small decrease in the red color pigment may be due to the polymeric components formed after the condensation reactions of tannins with phenolics during the long aging process (V3 sample) and the tannins losing their effect (A520, V2-1,336 and V3-1,193) on the red color pigment slightly. In addition, another phenolic component that is effective in color along with tannins in grapes and then wine is anthocyanins (Singleton 1969). It has been observed in studies that as the amount of anthocyanin decreases due to aging or vinification, there is a shift from red color to more yellow color (Almela 1993, Pascu 2005, Alcalde-Eon et al. 2006, Uysal et al. 2023). Moreover, it has been reported that the concentration of monomeric anthocyanins decreases during acetification process of vinegars because of polymerization reactions (Cerezo et al. 2010). Therefore, although the concentration of some phenolic compounds decreases, the slight change in color can be explained by the low level of anthocyanins. In a study, the browning measurement (absorbance at 420 nm) of traditional balsamic vinegar (TBV) was studied during the aging period and some colorless and yellowish compounds were determined because of phenolic reactions (Verzelloni et al. 2010). The formation of

colorless polymers in TBV has been explained by flavonoids polymerization reaction catalyzed by acetaldehyde. Besides, formation of new colored-yellowish oligomers and polymers has been also explained that glyoxylic acid might promote flavonoid polymerization (Es-Safi et al. 2002). In conclusion, in this study, as mentioned in the previous section, a decrease in the amount of some phenolic compounds (flavonoids and phenolic acids) was observed because of condensation and polymerization reactions, but the effect of these phenolic compounds on color change was not found to be statistically significant. It is evident that the aging period influences some phenolic compounds of vinegar samples, leading to transformations in their organoleptic properties. Notably, there is a lack of studies in the literature focusing on "Sherry" vinegar that simultaneously assess aging time, phenolic profile, and color properties. This highlights the potential for further research in this area to enhance our understanding of vinegar composition and quality.

Conclusion

In this study, color properties (pigments (%), color intensity, tonality, and density), phenolic compounds, and correlation of color properties-phenolics of *Sherry* vinegars during the aging process were examined. The results showed that the contents of phenolic compounds and color properties changed during the aging period of vinegar samples. The percentage of red pigment increased while the percentage of yellow pigment decreased during aging up to 12 years. It was revealed that a long period of aging in wood casks facilitated Maillard reactions resulting in a different pigmentation with browning compounds. Twelve phenolic compounds were identified, and a high negative correlation was observed between the content of total phenolic compounds and the aging period of vinegar samples. While the aging period of the samples increased, a decrease in the content of some phenolic compounds was observed. As a result, the aging period of Sherry vinegars caused a significant change from yellow to red tone, and a decrease in the amount of monomeric phenolic compounds. This study could serve as a catalyst for future research on Sherry vinegars, particularly in exploring other organoleptic properties and chemical profiles.

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