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Effect of ageing on lees and distillation process on fermented sugarcane molasses for the production of rum

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ABSTRACT

The study aimed at evaluating the influence of fermented sugarcane molasses ageing on lees and the distillation process used for the production of rums. Molasses were freshly fermented or 3-months lees aged. Batch (PS: Pot Still) or continuous (CS: Coffey Still) distillation was carried out resulting in four different rum distillates. Gas chromatography and 3D-fluorescence enabled to differentiate rum distillates chemical composition according to the distillation process, regardless of the ageing on lees of fermented molasses. Differences in fluorescent PARAFAC components and volatile acids, acetals and carbonyls contents revealed the predominance of the physicochemical processes driven at the liquid-vapor interface of fermented molasses, generated by the distillation systems. Notwithstanding the distilling conditions, the long chain fatty ester content was significantly higher in the 3-months lees aged condition. Multivariate analysis highlighted that CS rum distillates were chemically more homogeneous than those obtained by PS that preserved the lees effect.

1. Introduction

Sugarcane molasses are the viscous end product of sugar companies which is mostly valued as raw material prior to fermentation and distillation for rum production. The choice of yeasts and the conditions of fermentation differentiate molasses wort chemical composition which are revealed later in the characteristics of volatile composition of distillates (Medeiros et al., 2017). During the elaboration of fruit, cereal or plant fermented beverages, a great diversity of microorganisms can be used but the yeast *Saccharomyces cerevisiae* remains the main species generally used (Campos et al., 2010; Walker & Stewart, 2016). Additionally in the area of distilled beverages, particularly in whisky production, specific strains of *Saccharomyces cerevisiae* have been selected for their high alcohol content tolerance and their capacities to convert mash sugars into ethanol, carbon dioxide and numerous flavor congeners (Stewart, Hill, & Russell, 2013). In the area of rum production, the inoculation of selected yeasts strains for sugar cane fermentation can be excluded, in favor of the expression of indigenous microbial flora, often associated with rums richer in aromas. For example, the "Rhum Agricole" involves a complex indigenous microbiota made of mixes of yeasts and bacteria, already present in the sugarcane juices. *Lactobacillus* and *Propionibacterium* species have also been shown to remain in sugarcane molasses used for "Rhum Grand Arôme" production (Fahrasmane & Ganou-Parfait, 1998). Another practice used for producing heavy rums consists of adding the "dunder" in the fermenting molasses wort. The "dunder" is the residual creamy vinasse from the previous distillation, made of sugars and dead yeast cells (Fahrasmane & Parfait, 2003; Medeiros et al., 2017). Such ancestral practice could be hazardous with the risk of low alcoholic fermentation yields, unachieved fermentations and the development of spoilage microorganisms. The control of fermentation can be improved by direct inoculation of pure cultures of microorganisms or inoculation of a mother yeasting pre-cultured in a fermenter. In some cases, dried yeasts can be directly added in the washing media (Fahrasmane & Ganou-Parfait, 1998; Murtagh, 2003). The choice of strains impacts the quality of rums. The distinction between the different types of rums, light or heavy rums for instance, can be designed by the choice of inoculated yeast strains belonging to *Saccharomyces cerevisiae*, *Saccharomyces bayanus* or *Schizosaccharomyces pombe* (Fahrasmane & Ganou-Parfait, 1998; Medeiros et al., 2017). The quality of the final spirit can be also modulated with a sugarcane fermentation obtained by co-inoculation of a consortium of microorganisms (Duarte, de Sousa, Dias, & Schwan, 2011). Moreover, the presence of yeast lees in the mash could positively impact the spirit's quality, especially for heavy rums (Medeiros et al.,

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2017; Murtagh, 2003). The presence of yeast lees during distillation has been shown to promote different releases in ethyl esters, ethyl hexanoate and octanoate in particular, leading to differences in rum styles (Suomalainen, 1981).

Rum technology involves two distillation techniques used all around the world of distilled beverages: the ancestral one with the pot still and the industrial one with coffey still (Fahrasmane & Parfait, 2003). In both cases, odorous volatile compounds, concentrated in the final spirit, enabled a classification of the different types of rums according to their level of concentration. Traditional agricultural rums produced from raw sugar cane differ from sugar refinery molasses rums in composition and concentration, generally due to differences in the distillation process (Pigott, 2003). Liebich, Koenig, and Bayer (1970) identified more than 200 flavor compounds in a Jamaican rum using liquid extraction of rum prior to rum analysis by gas chromatography coupled to mass spectrometry, with concentrations reaching 800 ppm, particularly for fused alcohols (Liebich et al., 1970). According to Marse et al. (2004) rum is one of the distilled beverages that has the most volatile compounds, reaching 550 different aromas (Maarse & Van Den Berg, 1994). Some Grand Arôme and heavy rums, often appreciated from rum tasters due to their elevated esters content, can reach concentrations of more than 500 g/hL of pure alcohol (Fahrasmane & Ganou-Parfait, 1997). According to Fahrasmane and Ganou-Parfait (2011), the control of the organoleptic quality of heavy rums production remains a big challenge for rum producers and scientists due to the variability in microbiota and the impact of distillation processes This study presents a quantification of the effect of ageing on lees and the distillation process based on the quantification of chemical differences in the composition of major volatile compounds families and fluorescent components. The discrimination potential of each fermenting and distilling practices in sugarcane molasses rums was evaluated by multivariate statistical analysis.

2. Materials and methods

2.1. Wort samples and fermentations

2.1.1. Sugarcane molasses characteristics and wort preparations

Sugarcane molasses were supplied by a French rum company (Compagnie des Indes, Beaune, France). Prior to fermentation, the molasses were diluted with distilled water, in order to obtain 50 kg of diluted molasses characterized by a density of 1.090 at 20 °C with a DMA 35 densimeter (Anton Paar, Graz, Austria). The diluted molasses presented a Brix degree of 16 and an initial pH of 4.9. Then 16 kg of diluted molasses were poured into three 20 L glass demijohns and supplemented with 30 g/hL of diammonium phosphate (Sigma), 30 g/hL of yeast assimilable nitrogen (Mauriferm Gold, AB Maury, Peterborough, UK). The strain of *Saccharomyces cerevisiae* was Pinnacle MG+ (AB Mauri, Peterborough, UK), packaged in active dry form. The yeast inoculation was applied at the dose of 40 g/hL, according to the manufacturer's recommendations.

2.1.2. Fermentation processes

The fermentations were conducted in demijohns without stirring at room temperature (18–25 °C) and monitored in terms of density and temperature. Measures were realized twice per day with a DMA 35 densimeter (Anton Paar, Graz, Austria). Demijohns were weighed with a numeric analytical scale of 35 kg (Mettler Toledo, Greifensee, Switzerland).

Two series of fermentations were carried out in biological triplicates at three months of interval. After fermentation, the first mashes were left at 4 °C in contact with the yeast lees (L: Lees) to age during three months (*L1, L2, L3*).

The second fermentations (F: Fresh) were carried out in triplicates (*F1, F2, F3*), with the same protocol as previously described, just prior to the distillation. In all cases, yeast lees (fresh or aged) were removed from mashes before distillation.

S. *cerevisiae* strain implantations were controlled at the middle of alcoholic fermentation using a PCR interdelta analysis according to a previously published procedure (Legras & Karst, 2003). As illustrated in Fig. S.I.1 all sugarcane molasses were fermented with the same yeast strain.

2.2. Distillates samples

Two types of distillation: the pot still (*PS*) and the column still (*CS*) were carried on the six samples of fermented sugarcane molasses. Distillation systems used in this study can be viewed in Fig. S.I.2. For that, half of the demijohn content, corresponding to 8 kg was poured into the pot still and 8 kg was poured into the column still generating twelve distillates that were used for chemical analyses.

2.2.1. Pot still distillation

Pot still distillation was heated directly by flame contact with the copper surface of the 25 L copper still. Two distillations were carried out, the first one leading to the "low wines" and the second one leading to the final white distillated spirit. Volumes and ethanol content of these final distillates were analyzed. For this second pot still distillation, we decided to cut at 50% of alcohol content for the six wort batches (*PS-F1, PS-F2, PS-F3, PS-L1, PS-L2, PS-L3*) in order to keep an optimized control of pot still distillation process. The foreshots were removed and corresponded in each case to an approximated volume of 100 mL characterized by an intense solvent olfactive character.

2.2.2. Coffey still distillation

Column still distillation was carried out on a 25 L Holstein column (Markdorf, Germany). Temperatures in the boiler, heater, column and deflegmator were automatically measured, with a control of the boiler temperature. Heat was generated by a steam flow in direct contact with the copper still and controlled by a pressure of 150 mbars, enabling to keep a constant temperature of 90 °C inside the still. Temperature, alcohol content and distillate flow rate were automatically monitored online thanks to an infrared detector for the six wort batches (*CS-F1, CS-F2, CS-F3, CS-L1, CS-L2, CS-L3)*. The control of the cooling system was adjusted with an automatic valve. The distillate flow rate was kept constant between 15 and 20 mL/min and collected as the hearts of the distillation and once passing below 10 mL/min the hearts were separated from the tails. The foreshots were removed the same way as described in the pot still distillation

2.3. Chemical analysis

2.3.1. Wort and distillate characterization

Wort and distillate classical parameters such as ethanol, pH and total acidity and ethanol (only for distillates) were determined according to OIV standardized methods (Recueil des, 1994). Ethanol content was determined in the mashes at the end of fermentation by an enzymatic method following the manufacturer's instructions (Bio-Sentec®, France).

2.3.2. Distillate volatile composition

The distillates were also submitted to a targeted analysis of the volatile chemical composition. The liquid extracts (990 mL of distillate sample and 10 µL of octan-3-ol at 1 g/L) were analyzed with a Agilent Technology 5975C spectrometry (Shimadzu QP2010+, electronic impact at 70 eV) paired with a Agilent Technology 7890 A gas chromatograph fitted with a split/splitless injector (250 °C). The chromatograph was equipped with a capillary column PEG of 30 m × 0.32 mm (J &W Scientific). Film thickness was 0.50 µm. Helium was used as vector gas at a rate of 1.5 mL/min (average velocity of 44 cm/*sec*). The temperature of the oven was increased from 50 °C to 240 °C at 5 °C/min, and finally held at 240 °C for 5 min. The injection mode was splitless.

The analyses were done in triplicate. Spectrometry Selected Ion Monitoring method (SIM method) was used for molecules detection. The mass spectrometer scanned from *m/z* 29 to 500. The volatile compounds were identified by matching their spectral fragmentation with those provided by the mass spectral library of the National Institute of Standards and Technology (NIST) and the Wiley Registry (WILEY) and by validating with pure chemical standards. Quantification was carried out via an internal standard method by the addition of octan-3-ol to distillates reduced to 50% ethanol (v/v) with ultrapure water prior to injection Response factors were calculated for volatile compounds from calibration curves obtained by analyzing hydroalcoholic solutions (ethanol 50%, v/v) made from pure analytical grade standards (SigmaAldrich, Saint Louis, MO) in the ranges 0.05–10 mg/L for phenylethanol, eugenol, ethyl acetate, isoamyl acetate, ethyl lactate, ethyl butanoate, ethyl octanoate, ethyl decanoate, ethyl dodecanoate, 1,1-diethoxy ethane, diacetyle, 2-methyltetrahydrofuran-3-one, furfural, propanoic acid, *n*-decanoic acid, propanoic acid, 2-methyl and octanoic acid and 1–200 mg/L for propanol, 2-methyl-propanol, butanol, 3-methyl-butanol, 2 methyl-butanol. The concentrations of volatile compounds were converted in grams per hectoliter of pure alcohol following CE regulation 2870/2000.

2.3.3. Excitation Emission Matrices of fluorescence (EEMF) of rum distillates

All rum distillates were analyzed with an untargeted approach consisting of measuring Excitation Emission Matrices of Fluorescence (EEMF). For that, rum distillates were diluted twenty times with ultrapure water and put in 1 cm path-length quartz cuvette and EEMFs were recorded in a Horiba Aqualog unit, enabling to automatically correct the Rayleigh and Raman scattering and the inner filtering effect and to normalize EEMFs to a quinine sulfate 1 ppm solution.

2.4. Statistical analysis

Aroma concentrations were statistically analyzed by multivariate analysis using Origin Lab software. PARAFAC model of rum distillates EEMFs was built on home made Matlab software, previously used for wine PARAFAC modeling (Coelho et al., 2015). PARAFAC model was validated by core consistency and split half validation of the dataset. PARAFAC model described each PARAFAC components by their fluorescence intensites at their maximum, represented as Fmax values. Fmax values were used to statistically interpret the distillate fluorescent composition and classify the different rum distillates in function of their elaboration processes.

Mean Fmax values of PARAFAC components and mean volatile compounds concentrations were statistically compared with an ANOVA test with an interval of confidence of 95%, followed by a Tukey's HSD post hoc test to evaluate the impact of yeast lees ageing and distillation practice.

3. Results and discussion

3.1. Fermentation monitoring

The evolutions of the molasses wort density, weight and temperature upon the fermentation stage for the fresh (F) and yeast lees (L) modalities are presented in Fig. 1A and B, respectively. Fermentations started at a density of 1.086 and reached a final density of 1.030 for each modality. Molasses weight decreased from 16.1 kg to 15.3 kg for F modalities and from 16.0 kg to 15.1–15.2 kg for L modalities. For F modalities, fermentation started just after the yeast strain inoculation and finished within 48 h for the three biological replicates (F1, F2 and F3). For L modalities, we observed a lag phase of 24 h following yeast strain inoculation for L1 and L2. This lag phase was around 40 h for L3. These delays were probably due to lower non-controlled fermentation temperatures of 20 °C compared to 26 °C for the fresh modality. Fermentation at lower temperatures values affected yeast metabolism by slowing their proliferation in the molasses wort. Nevertheless, the real duration of the alcoholic fermentation for L modalities was comparable to that obtained with F modalities, *ie* 48 h. Ethanol contents measured at the end of the alcoholic fermentation are specified in Fig. S.I.3. For all modalities, the average ethanol contents presented no statistical differences ($p = 0.05$) and were comprised between 6.45% and 6.80%, for L and F modalities, respectively. Final pH was measured at 4.5 and 4.6 for (F) and (L) conditions, respectively.

3.2. Rum distillates chemical analysis

3.2.1. Volatile congeners composition

Major volatile congeners concentrations were quantified in each rum distillate. Fig. 2A illustrates a heatmap representation of volatile compounds normalized by the maximum concentration found among the twelve samples per volatile compound and grouped by chemical families (alcohols, esters, acetals, carbonyls and acids). The mean concentrations for CS and PS rum distillates, regardless of the presence/absence of lees on fermented sugarcane molasses are presented in Fig. 2B. The mean concentrations for L and F rum distillates, regardless of the distillation process are indicated in Fig. 2C. Raw concentrations values of individual volatile congeners found in rum distillates are indicated in additional information (Fig. S.I.4).

3.2.1.1. Distillation process differentiation. First of all, CS and PS rum distillates generated by the two distillation systems presented different normalized concentrations of volatile congeners, particularly for chemical families like acetals, carbonyls and acids and to a lesser extent alcohols (Fig. 2A). Statistical differences were found in PS distillates with higher concentrations in acetals (1,1-diethoxyethane), carbonyls (furfural, diacetyl, 2-methyloxolan-3-one) and acids (propanoic, isobutyric, octanoic, decanoic) compared to CS distillates. No statistical differences were found for alcohols and esters (Fig. 2B). Were also more detected in PS rum distillate, some individual volatile compounds such as 3-methyl-propanol phenylethanol, eugenol, ethyl acetate and ethyl lactate (Fig. S.I.4). Such results have already been pointed out in brandy, cachaça and whisky production (Maarse & Van Den Berg, 1994; Nascimento, Cardoso, & Franco, 2008; Piggott & Paterson, 1994; Simpson, 1971). Furfural, already present in sugarcane molasses, is formed by Maillard reaction when using direct heating pot still units (Simpson, 1971). To our knowledge methyloxolan-3-one, a Maillard reaction product already found in rum (Nykänen & Suomalainen, 1983) which has a pleasant coffee note has never been shown to depend on the type of distillation. 1,1-diethoxyethane, conferring a fruity note to the distillate was only present on pot still batches and was not detected in the CS rum distillates, meaning the continuous distillation reduced acetals formation (Piggott & Paterson, 1994). Organic acids were not detected in CS distillates, revealing they were eliminated due to different partitioning of these compounds in the CS column plates, particularly due to the elevated amount of reflux (Maarse & Van Den Berg, 1994). Another plausible reason is these organic acids were more prone to esterification with ethanol leading to higher concentrations of their esterified forms, particularly ethyl hexanoate, ethyl octanoate, ethyl decanoate and ethyl dodecanoate.

3.2.1.2. Lees ageing effect after distillation. Interestingly, ester compounds were more present in rum distillates generated from yeast lees aged mashes compared to the fresh mashes , independently of the distillation process (Fig. 2A and Fig. 2C). This increase in ester content in rum distillate had already been described when lees were directly incorporated into the still with a progressive release of their lipophilic content in the wort with the temperature increase during the distillation Suomalainen (1981). This abundance in ester compounds was never previously attributed to the lees ageing process on fermented sugarcane molasses. Only the 3-months yeast lees aged rum distillates

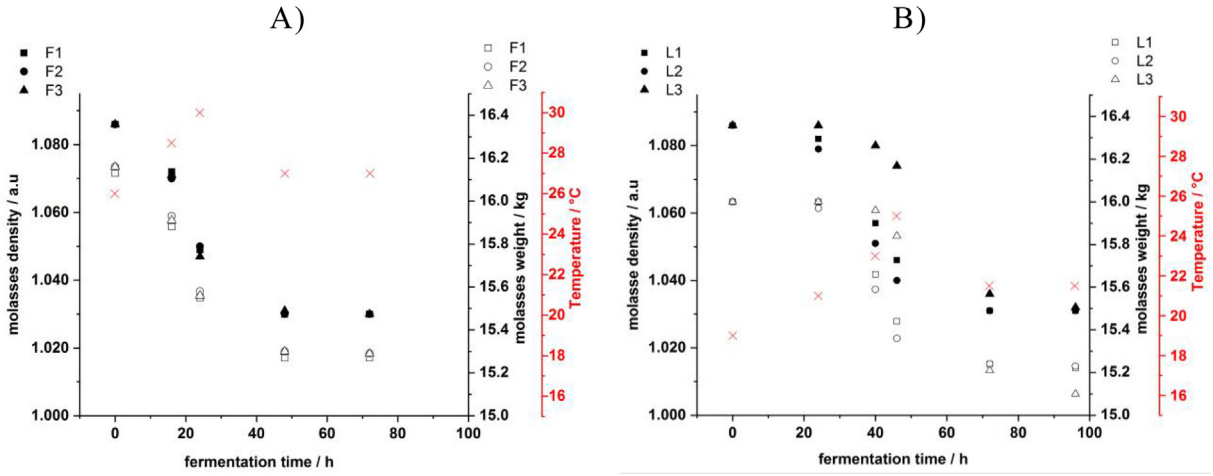

Fig. 1. Fermentation monitoring of sugar cane molasses wort density (filled symbols), weight (emptied symbols) and temperature (red cross) for the three biological replicates for (A) fresh (F1, F2 and F3) and (B) 3-months yeast lees aged (L1, L2 and L3) modalities.

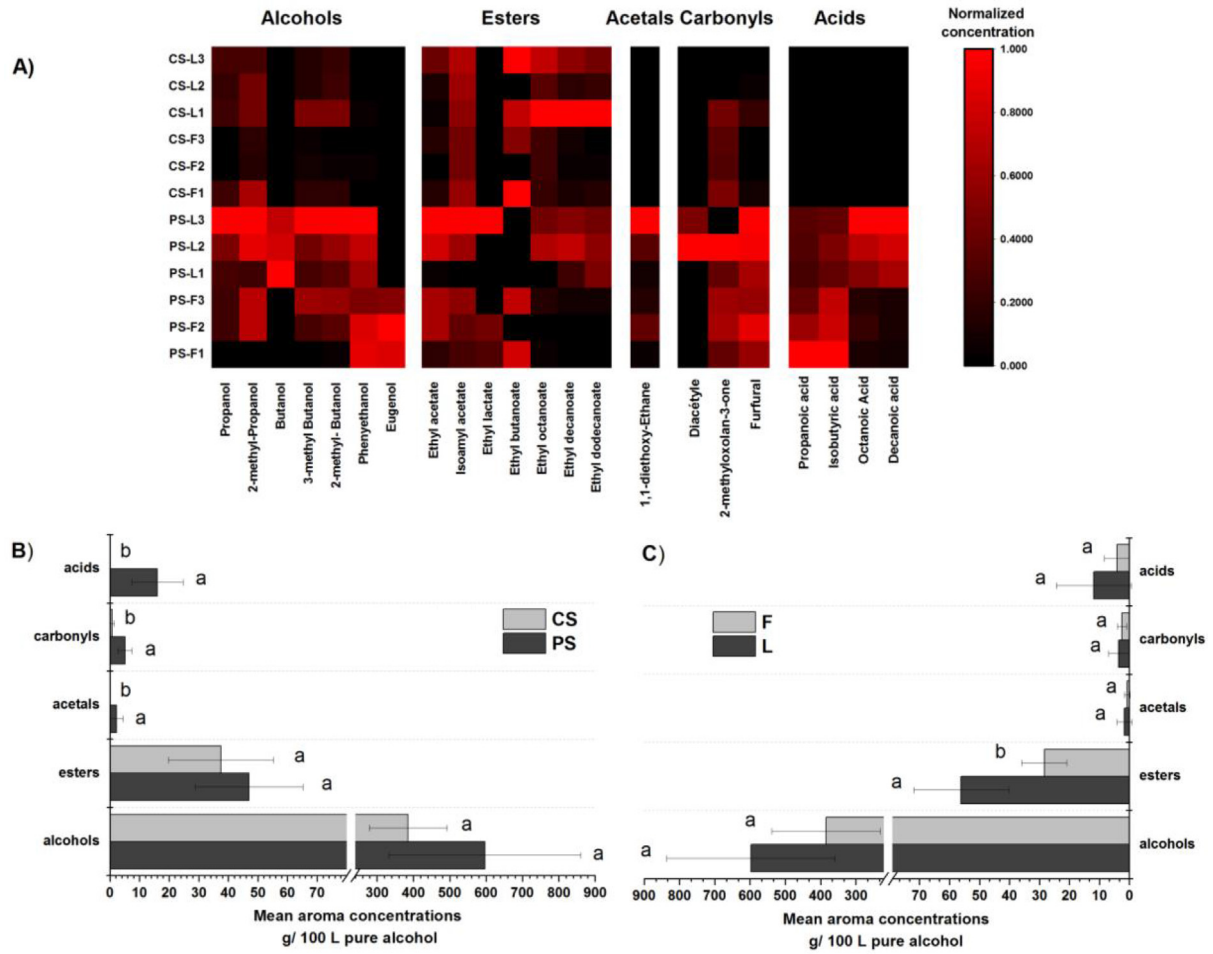

Fig. 2. (A) Heatmap of aroma compounds quantified in the rum distillates after Coffey still (CS) and Pot still (PS) distillations from fresh (F) and 3-months yeast lees aged (L) sugarcane molasses fermentations in triplicates. Concentrations are normalized by the maximum concentration per volatile congeners and represented by the color scale from black (O) to red (1). Mean aroma concentrations grouped by chemical families by comparing PS and CS distillation regardless of the type of fermentation (B) and by comparing L and F fermentation regardless of the type of distillation (C). Letters a and b indicate the results of the variance analysis performed for each chemical family.

showed higher amounts of 1,1-diethoxyethane, diacetyl, octanoic acid and decanoic acid compared to the fresh rum distillates (Fig. S.I.4). Such fatty acids increase after 3-months lees aging has been proposed by Troton et al. (1989) as degradation of membrane compounds from cells (Troton, Charpentier, Robillard, Calvayrac, & Duteurtre, 1989). Nevertheless, propanoic and isobutyric acids were found in higher amounts in fresh fermented rum distillates traducing their preferential accumulation in the distillate after a pot still distillation. The same

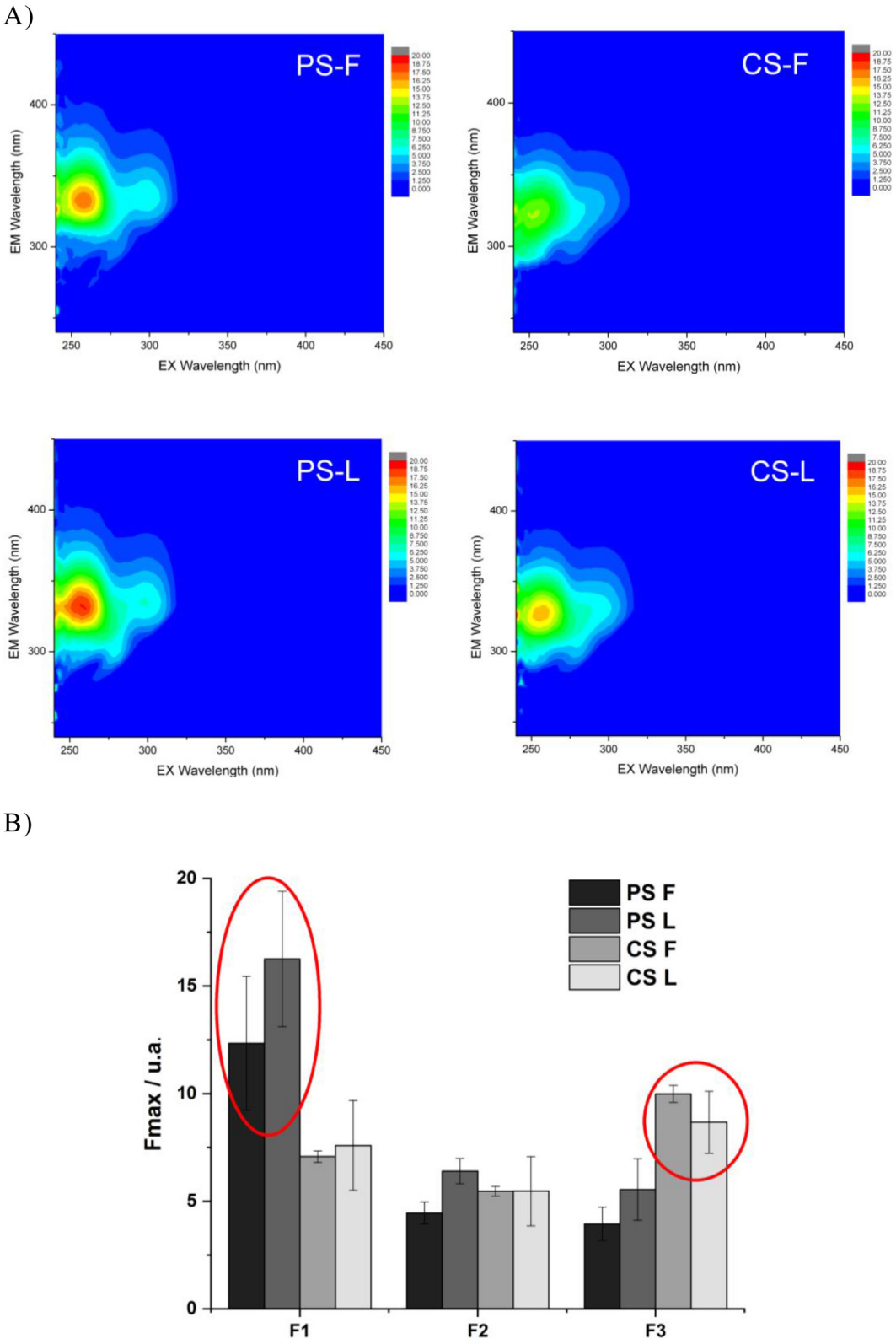

Fig. 3. Excitation Emission Matrices of Fluorescence of the four rum distillates PS-F, CS-F, PS-L and CS-L (A) and mean Fmax values of PARAFAC components F1, F2, and F3 of the same four rum distillates analyzed in biological triplicates (B).

tendency is observed with compounds like eugenol or furfural, which are more present in fresh fermented rum distillates. As previously mentioned in wine medium, these woody-flavored compounds tend to bind to yeast lees and be less detected in the resulting wines (Chatonnet et al., 1992; Jiménez Moreno & Ancín Azpilicueta, 2007). This phenomenon could also explain the reduced concentration of eugenol and furfural in the rum distillates from 3 months lees aged mashes.

3.2.2. *Rum distillates EEMF analysis*

The chemical composition of rum distillate was assessed by means

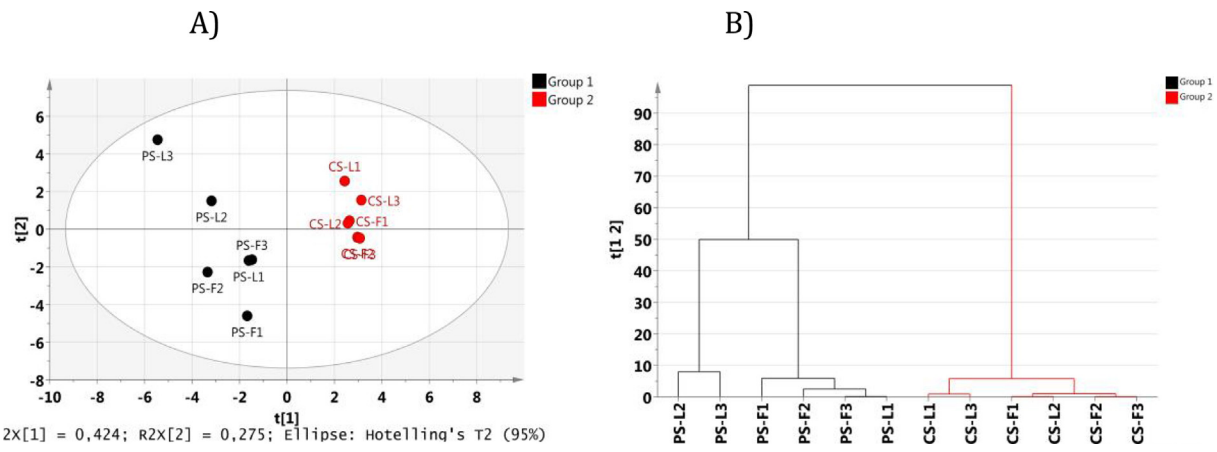

Fig. 4. Statistical discrimination of rum distillates based on their chemical analysis and the way fermentation and distillation was carried measured by a partial least squares discrimination analysis (A) and a hierarchical clustering analysis (B).

of 3D fluorescence spectroscopy in order to strengthen the previous volatile congeners differentiations between the fermentation and distillation modalities. Excitation-Emission Matrices of Fluorescence of rum distillates elaborated from fresh and 3-months yeast lees aged sugarcane molasses in pot still and coffey still are shown in Fig. 3A. All rum distillates present two typical emission areas centered at 340 nm for 250 and 300 nm of excitation wavelengths. These emissions have been attributed in other food systems to a great variety of compounds such as phenolics, furfurals, NADH and Maillard reaction products (Coelho et al., 2015; Elcoroaristizabal et al., 2016; Ghosh, Verma, Majumder, & Gupta, 2005; Markechova, Majek, & Sadecka, 2014; Matiacevich & Pilar Buera, 2005). The intensity of each emission area was higher in rum distillates from pot still compared to coffey still, regardless of the lees ageing on mashes. For finest discriminations and statistical validation, a PARAFAC model was built based on the analysis of twelve rum distillates samples analyzed in triplicates. The model generated three PARAFAC components (F1, F2 and F3), shown in Fig. S.I.5, enabled to statistically differentiate the effect of distillation process used in the elaboration of rum distillates. Fig. 3B illustrates this differentiation obtained by analyzing each Fmax values of the model. PS rum distillates present higher mean Fmax values of PARAFAC component F1 from 12.34 (PS F) to 16.25 (PS L) compared to CS rum distillates (Fmax mean values of 7.07 and 7.59, for CS F and CS L, respectively). CS rum distillates present higher mean Fmax values of PARAFAC component F3 from 9.99 (CS F) to 8.67 (CS L) compared to PS rum distillates (Fmax 3 mean values of 3.95 and 5.54 for PS F and PS L, respectively. No statistical differences were found for Fmax values of PARAFAC component F2 for the four rum distillates. This spectral discrimination between batch and continuous distilled liquids by means of PARAFAC components F1 and F3 could be attributed to the influence of volatile compounds mainly present in distillates such as alcohols, esters and acids that affect the chemical environment of intrinsic fluorophores (Sadecka, Urickova, Jakubikova, 2016). Longer wavelength emissive compounds, associated to the statistical PARAFAC component F1, could also be attributed to volatile carbonyls such as furfural, that were analytically measured at higher levels in PS distillates (previously shown in Fig. 2B), coinciding with the observed higher Fmax values of this component. Nevertheless, chemical assignments should be performed carefully due to several overlapping bands originating from different volatile fluorophores present in the total fluorescence spectra of rum distillates.

3.3. Impact of the lees ageing and distillation practices

As rum distillates were differentiated by means of their volatile congener composition and their fluorescence fingerprinting, prediction statistic models were built using multivariate approaches by partial least squares discrimination analysis and hierarchical clustering analysis. Results are shown in Fig. 4 where volatile congeners concentrations and PARAFAC components were used as predicable variables and the distillation type (PS: group 1 or CS: group 2) as dependent variables. Fig. 4A illustrates statistically the clear discrimination found between the two types of distillation along the first component t[1] regardless of the treatment of mashes after fermentation. This PS/CS distinction is essentially driven by higher Fmax values of PARAFAC component F3 and some long chain fatty esters in C8, C10 and C12 for CS rum distillates and by higher values in Fmax 1, volatile acids, furfural and phenylethanol in PS rum distillates. Fig. 4B shows the number of clusters and the level of cluster similarity represented by the Y-axis. It is interesting to notice that CS distillates presented closest similarities compared to the PS rum distillates independently of fermented mashes. In the same way, PS rum distillates presented close similarities once they were elaborated from fresh fermented sugarcane molasses whereas the 3-months yeast lees aged one led to a higher discrepancy between the triplicates of rum distillates. This statistical approach permitted a better evaluation of the variability of the distillation process taking into account the heterogeneity of fermented sugarcane molasses. Continuous distillation enabled a better homogenization of rum distillates whereas batch distillation preserved the yeast lees ageing practice on mashes that could be applied or desired by some rum producers.

4. Conclusion

Sugarcane molasses were fermented freshly or yeast lees aged during three months prior to distillation in order to obtain different styles of rum distillates. Regardless of the nature of the distillation process, yeast lees ageing led to higher amounts of ester contents, particularly long chain fatty esters and some of their precursors like fatty acid in C8 and C10. Once distillation is carried out, pot still rum distillates differ from coffey still distillates by presenting specific fluorescence fingerprinting related to their chemical volatile composition. This study also highlights for the first time that yeast lees ageing practice on sugarcane molasses coupled to batch distillation could confer a differentiated rum style whereas continuous distillation tends to minimize its impact.

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.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.foodchem.2019.125405.

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