Food Chemistry 229 (2017) 743–751

Contents lists available at ScienceDirect

Food Chemistry

journal homepage: www.elsevier.com/locate/foodchem

Differentiation of Chinese robusta coffees according to species, using a combined electronic nose and tongue, with the aid of chemometrics

Wenjiang Dong [a,b,c], Jianping Zhao [a,b,c], Rongsuo Hu [a,b], Yunping Dong [a,c], Lehe Tan [a,b,c,*]

[a] Spice and Beverage Research Institute, Chinese Academy of Tropical Agricultural Sciences, Wanning, Hainan 571533, China
[b] National Center of Important Tropical Crops Engineering and Technology Research, Wanning, Hainan 571533, China
[c] Key Laboratory of Genetic Resources Utilization of Spice and Beverage Crops, Ministry of Agriculture, Wanning, Hainan 571533, China

ARTICLE INFO

Article history:
Received 6 October 2016
Received in revised form 18 February 2017
Accepted 28 February 2017
Available online 2 March 2017

Keywords:
Coffee
Electronic nose
Electronic tongue
Data fusion
Chemometrics
Classification

ABSTRACT

Electronic nose and tongue sensors and chemometric multivariate analysis were applied to characterize and classify 7 Chinese robusta coffee cultivars with different roasting degrees. Analytical data were obtained from 126 samples of roasted coffee beans distributed in the Hainan Province of China. Physicochemical qualities, such as the pH, titratable acidity (TA), total soluble solids (TSS), total solids (TS), and TSS/TA ratio, were determined by wet chemistry methods. Data fusion strategies were investigated to improve the performance of models relative to the performance of a single technique. Clear classification of all the studied coffee samples was achieved by principal component analysis, K-nearest neighbour analysis, partial least squares discriminant analysis, and a back-propagation artificial neural network. Quantitative models were established between the sensor responses and the reference physicochemical qualities, using partial least squares regression (PLSR). The PLSR model with a fusion data set was considered the best model for determining the quality parameters.

1. Introduction

Coffee is one of the most popular and widely consumed beverages in the world and exerts strong economic and cultural influences. The most important species of coffee are *Coffea canephora* (robusta) and *C. arabica* (arabica) (Monakhova et al., 2015). In 2012, the total economic value of the coffee industry was approximately $173.4 billion, and global coffee consumption has risen at an average annual rate of 1.9% over the past 50 years (Lee, Cheong, Curran, Yu, & Liu, 2015). All these phenomena can be ascribed to the attractive and pleasant aroma and taste of coffee (Petisca, Pérez-Palacios, Farah, Pinho, & Ferreira, 2013; Sunarharum, Williams, & Smyth, 2014). Coffee production mainly occurs in Southern and Central America, the Caribbean countries, Africa, and Asia. Coffee contains a large number of antioxidant compounds, caffeine, trigonelline, soluble solids, and a few secondary metabolites, such as minor chlorogenic acids isomers, organic acids, and diterpenes. Coffee has antioxidant (Ramalakshmi, Kubra, & Jagan Mohan Rao, 2008), anti-carcinogenic (Giovannucci, 1998), and anti-mutagenic activity (Kim & Levin, 1988).

In the past 10 years, coffee has gained acceptance in China, owing to increased communication between the East and West, and its consumption has increased by 4-fold in the last 10 years (Tie, Hu, & Zhang, 2016). Coffee production in China mainly occurs in the Hainan and Yunnan Provinces, and the most commonly planted variety in the Hainan Province is robusta coffee. Xinglong and Fushan coffees are the most popular brands in the Hainan Province, possess peculiar traits and unique qualities, and are protected by the Chinese General Administration of Quality Supervision, Inspection and Quarantine with protected designation of origin status (Ouyang, Wang, Long, Dong, & Fu, 2012). The quality of coffee is closely related to the roasting process. At present, the 3 main coffee roast degrees are the light degree (e.g., light city, half city, cinnamon roast, and New England roast), the medium degree (e.g., regular roast, American roast, city roast, and breakfast roast), and the dark degree (e.g., French roast, Italian roast, espresso roast, continental roast, New Orleans roast, and Spanish roast) (Kučera, Papoušek, Kurka, Barták, & Bednář, 2016). Distinguishing coffee cultivars is essential for farming and plant germplasm resource preservation because different cultivars show variations in flavour and taste.

Several studies have been conducted to evaluate the quality of coffee and gauge its authenticity over the past few years

* Corresponding author at: Spice and Beverage Research Institute, Chinese Academy of Tropical Agricultural Sciences, Wanning, Hainan 571533, China.
E-mail addresses: dongwenjiang.123@163.com (W. Dong), zjp-68068@163.com (J. Zhao), hnhrs@126.com (R. Hu), 437433465@qq.com (Y. Dong), tlh3687@163.com (L. Tan).

http://dx.doi.org/10.1016/j.foodchem.2017.02.149
0308-8146/

(Garrett et al., 2013; Sberveglieri et al., 2014; Várvölgyi et al., 2015). Both the commercial classification and the evaluation of the authenticity of coffee depend on sensory panels. However, sensory evaluation is a time-consuming process with low objectivity and reproducibility. Furthermore, flavour assessment by a human is inevitably associated with problems involving the standardisation of measurements; the stability of the evaluation; and the physical condition, health, and mood of the panellists (Qiu, Wang, Tang, & Du, 2015). Authentication of coffee samples from different sources has been performed using several modern analytical techniques, such as element and stable-isotope profiling (Carter, Yates, & Tinggi, 2015), determining the content of bioactive compounds (Mehari et al., 2016) by high-performance liquid chromatography (HPLC)-mass spectrometry or gas chromatography-mass spectrometry (Dong, Tan, Zhao, Hu, & Lu, 2015; Jumhawan, Yusianto, Marwan, Bamba, & Fukusaki, 2013), and DNA fingerprinting (Nganou et al., 2012). However, these methods are expensive, time consuming, and require complex sample preparation and toxic organic reagents. Therefore, other techniques, such as combinations of ultraviolet-visible-near infrared spectroscopy, Fourier transform infrared spectroscopy, and ^{1}H nuclear magnetic resonance spectroscopy, have been utilised to determine the flavour qualities of coffee samples based on their spectral characteristics (Bertone, Venturello, Giraudo, Pellegrino, & Geobaldo, 2016; Kwon et al., 2015; Liang, Lu, Hu, & Kitts, 2016). However, most of these techniques also require highly skilled technicians and are costly. Thus, low-cost, fast, accurate, reliable, and robust analytical techniques are urgently needed to authenticate and predict the flavour qualities of coffee.

Electronic nose (E-nose) and electronic tongue (E-tongue) analyses are essential for the rapid detection of chemical components and sensory attributes. These devices mimic the human senses of odour and taste. Furthermore, these devices enable the gathering of global information on the analysed solution, including sensory data and analytical properties, which can be related through appropriate chemometric methods such as pattern recognition techniques and multivariate calibration methods. Successful use of the E-nose and E-tongue to differentiate the geographical origin or variety of agricultural products is well-documented for products such as coffee, tea, wine, fish, and meat (Cole, Spulber, & Gardner, 2015; Ghasemi-Varnamkhasti & Aghbashlo, 2014; Ha et al., 2015). Lopetcharat et al. (2016) proposed a new approach that relates the E-tongue to human perception to determine the quality of coffee. Generalised Procrustes analysis (GPA) showed a strong correlation between the E-tongue and human perception, and differences between numerous coffee samples from various sources were identified using this approach. Buratti, Sinelli, Bertone, Venturello, and Casiraghi (2015) distinguished between washed arabica, natural arabica, and robusta coffee using the E-nose and E-tongue in combination with principal component analysis (PCA). Severini, Ricci, Marone, Derossi, and de Pilli (2015) utilised the E-nose system and described changes in the aromatic profile of espresso coffee as a function of the grinding grade and extraction time. An electronic panel formed by an E-nose and E-tongue were applied to evaluate the oxygen exposure levels and polyphenolic contents of red wines (Rodriguez-Mendez et al., 2014). Moreover, Han, Huang, Teye, Gu, and Gu (2014) developed a novel method for detecting fish freshness by analysing E-nose and E-tongue data with chemometric methods, and satisfactory results were obtained through this method.

The combined use of electronic noses and tongues can be employed to assess food authenticity and food adulteration (Peris & Escuder-Gilabert, 2016). Although several studies have used E-nose and E-tongue sensors to classify the varietal and geographical origin of coffee samples and to determine the physicochemical parameters of coffee brews, the E-nose and E-tongue techniques have not yet been utilised to distinguish varietal origins or predict the flavour quality of robusta coffee beans in Xinglong region of Hainan, China.

The aim of this study was to investigate the use of E-nose and E-tongue sensors for differentiating coffee samples produced from 7 cultivars with 3 different roasting degrees in the Xinglong region. Classification was performed using pattern recognition techniques, PCA, the K-nearest neighbour (KNN) method, partial least squares discriminant analysis (PLS-DA), and a back propagation artificial neural network (BP-ANN), in order to estimate the pH, titratable acidity (TA), total solids (TS), total soluble solids (TSS), and the TSS/TA ratio. This study was also conducted to evaluate whether the use of instruments combined with a multivariate regression method, such as partial least squares regression (PLSR), could increase the accuracy of the built regression models.

2. Materials and methods

2.1. Chemicals and reagents

HPLC-grade methanol, glacial acetic acid, and formic acid were purchased form Merck KGaA (Darmstadt, Germany). Analytical-grade sodium hydroxide, potassium dihydrogen phosphate, hydrochloric acid, and phenolphthalein were acquired from Damao Chemical Reagent Factory (Tianjin, China), and potassium chloride was supplied by Guangzhou Xilong Chemical Ind., Co., Ltd. (Guangzhou, China). All chemicals and reagents were analytical or HPLC grade, and all solutions were prepared using deionised water from a Milli-Q system (Millipore, Hetai, Shanghai, China).

2.2. Samples and sample preparation

The green coffee (*C. robusta*) used in this study was harvested in 2014/2015 at the experimental base of the Spice and Beverage Research Institute of the Chinese Academy of Tropical Agricultural Sciences in Hainan, China. Fresh and healthy fruits were selected for this study at approximately the red ripeness stage. Seven cultivars of *C. canephora* (robusta coffee) were included and named as 'Robusta Xinglong 1' (X1), 'Robusta Reyan 1' (RY1), 'Robusta Reyan 2' (RY2), 'Robusta Xinglong 24-2' (X24-2), 'Robusta Xinglong 26' (X26), 'Robusta Xinglong 28' (X28), and 'Robusta Chenmai' (XCM). Three representative samples from each cultivar were used; thus, a total of 21 green coffee beans was included in this study. The fresh coffee fruits were mechanically hulled after harvesting, and the mucilage was removed. Coffee endosperms with parchment were subjected to hot air drying in an oven (Model 101-2-BS; Shanghai Yuejin Medical Instrument Co. Ltd., Shanghai, China) at 50 °C until the moisture content was approximately 10 11%.

Green coffee beans were roasted at constant power input in a PRE 1 Z coffee roaster with a rotating drum (Emmerich am Rhein, Germany). The initial roasting temperature was 180 °C, and samples (100.0 g) were roasted for 3 different times (8, 10, and 12 min) to obtain different roasting degrees (i.e., light, medium, and dark). Samples were rapidly cooled to room temperature. To obtain a representative set of coffee samples, all roasts were performed in 2 independent experiments. A total of 126 roasted coffee samples (21 samples × 3 roasting times × 2 roasting process replicates) was obtained. Before analysis, all roasted coffee samples were ground using a commercial coffee grinder (Kenia, Mahlkönig, Germany), and medium millings of ground samples were sieved and used for further analysis. All samples were stored vacuum-sealed in bags at 4 °C. The extracts for analysis of quality parameters were obtained by combining 8.25 g coffee powder and 150 mL deionised water at 95 °C in a 200-mL flask, stirring for 5 min,