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Detection of adulteration in cherry tomato juices based on electronic nose and tongue: Comparison of different data fusion approaches

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ABSTRACT

Seven approaches were employed for authentication of fresh cherry tomato juices adulterated with different levels of overripe tomato juices: 0–30%. Two e-nose measurements were considered, and the result indicates that a pretreatment of using desiccant prior to e-nose measurement is unnecessary. Principle Component Analysis (PCA), factor *F* and stepwise selection were applied for feature construction of fusion datasets. Qualitative recognition of adulteration levels was mainly performed by Canonical Discriminant Analysis (CDA) and Library Support Vector Machines (Lib-SVM). Quantitative calibration with respect to pH and soluble solids content (SSC) was performed using Principle Components Regression (PCR). All the approaches presented well classification performances, and prediction performances based on fusion approaches are better than based on sole usage of e-nose or e-tongue; yet classification and prediction performances based on different fusion approaches vary. This study indicates that simultaneous utilization of both instruments would guarantee a better performance than individually utilization of e-nose or e-tongue when proper data fusion approaches are used.

1. Introduction

Freshly squeezed fruit juices labeled as 100% fruit are 100% made of fresh fruits being most of them not processed or processed by means of novel techniques such as HPP (High Pressure Pasteurization) which preserves the overall freshness of the product and its organoleptic and nutritional characteristics (Faria et al., 2013). Fruits are relatively easy to authenticate by their morphological characteristics and flavor. The contents of sugars in fruit juices, combined with their acidity and aromatic profile, promote the exotic and characteristic flavors of each fruit (De Carvalho et al., 2008). However, the act of processing fruits into juices gives rise to the possibility of adulteration. Substitution of material with cheaper alternatives, i.e. addition of water, sugar, pulpwash, senescent fruits or economical substitutes are known topics for fruit juice issues (Reinhard et al., 2008). Traditionally, sensory evaluation and chromatographic techniques have been used to determine food quality. Sensory evaluation provides immediate flavor information but suffers from some disadvantages, namely the correctness of training, standardization of measurements, reproducibility, high cost and taste saturation of the panelist (Beullens et al., 2008). Chromatographic techniques such as gas chromatography–mass spectrometry (GC–MS), high performance liquid chromatography (HPLC) and ion chromatography are time-consuming and expensive, and they require skilled personnel to operate the equipment and interpret the analytical results (Baldwin et al., 1998).

Electronic nose (e-nose) and electronic tongue (e-tongue), which are actually simulations of human nose and taste bud, respectively, have proven to be a good alternative for traditional techniques in odor and taste analysis of food (Escuder-Gilabert and Peris, 2010). In the area of fruits and beverage, e-nose has been successful in monitoring the aroma of melons (Benady et al., 1995), pears (Oshita et al., 2000), apples (Saevels et al., 2003), wines (García et al., 2006), peaches (Zhang et al., 2008) and various fruit juices (Farnworth et al., 2002; Gobbi et al., 2010; Karlshøj et al., 2007; Reinhard et al., 2008), and e-tongue has also been successful in evaluating the taste of food covering the area of process monitoring (Turner et al., 2003), freshness evaluation and shelf-life investigation (Gomez et al., 2008), authenticity assessment (Parra et al., 2006), foodstuff recognition (Legin et al., 1997), quantitative analysis and other quality control studies (Beullens et al., 2006).

It is noticeable that the two sensor systems do not look at the same features when applied to the same liquid sample. The e-nose sensors are in contact with its headspace while the e-tongue electrodes are immersed in the sample (Di Natale et al., 2000). In several applications (Buratti et al., 2007; Cosio et al., 2007; Gil-Sánchez et al., 2011; Rong et al., 2000; Tudu et al., 2012; Winquist et al., 1999), simultaneous applications of the two sensor systems have demonstrated improvement when compared to individual utilization; yet some researchers (Cole et al., 2011) found that little contribution was made by e-nose when their self-developed

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combined system was employed for complex samples and different solutions. Meanwhile, after reading the more recent articles concerning fusion of e-nose and e-tongue, we found that in most cases (Apetrei et al., 2012; Cole et al., 2011; Gil-Sánchez et al., 2011; Masnan et al., 2012; Prieto et al., 2011; Tudu et al., 2012; Zakaria et al., 2010), the sensors of e-nose and e-tongue were simply concatenated. In addition, the combined system was mostly used for classification tasks, while the other fundamental data analysis task – prediction of related quality indices – was not often emphasized.

In this paper, we describe the use of an e-nose, an e-tongue and four fusion approaches using both of the e-nose and e-tongue instruments to discriminate adulteration in cherry tomato juices, as well as to predict the pH and soluble solids content (SSC) of the adulterated juice samples. The problems of sensor drift and humidity were also taken into consideration. Different feature extraction and sensor fusion approaches were discussed. The main objective of this research are: (1) To explore if employing anhydrous sodium carbonate as desiccant would improve the performance of e-nose when detecting liquid samples; (2) To compare the performances of fusion approaches based on different feature exaction and construction methods; (3) To explore if the simultaneous utilization of perceptual knowledge from both the instruments will increase the extent of information regarding the sample, or on the contrary, will they lead to data redundancy.

2. Materials and methods

2.1. Sample preparation

Chinese variety, *youbei* cherry tomatoes were picked twice for self-made tomato juice at the experimental orchard in Department of Horticulture, Zhejiang University, Hangzhou, China. The first picking time was on 7th, June, 2012, and the second time was on 11th, June, 2012. All the cherry tomatoes were picked at roughly red ripeness stage (more than 90% of the surface, in the aggregate, shows red color) (Agriculture, 1997) with approximately uniform size. The ones that were picked on the 7th were stored for 4 days at ambient atmosphere, 25 ± 1 °C and 80 ± 5% relative humidity, to become overripe with flesh softening. Juices of these cherry tomatoes were used as filler juices. The ones that were picked on the 11th were used for freshly squeezed juices. During juicing process, cherry tomatoes were placed in a fruit squeezer and juiced for 30 s. Fresh and overripe tomatoes were squeezed separately to make fresh tomato juices and filler juices, respectively. Fresh tomato juices were then blended with overripe tomato juices at four levels of adulteration from 0% to 30% (w/w) in steps of 10%, which can be of great practical interest. The four groups were: 0% (100% fresh tomato juice), 10% (90 g of fresh tomato juice adulterated with 10 g of overripe tomato juice), 20% (80 g of fresh tomato juice adulterated with 20 g of overripe tomato juice) and 30% (70 g of fresh tomato juice adulterated with 30 g of overripe tomato juice). After blending, juices were filtered using medical gauze that was folded into eight layers. Filter liquor was collected for later e-nose and e-tongue detection.

2.2. E-nose and e-tongue instruments

Headspace analysis was performed with a PEN2 e-nose (Airsense Analytics, GmBH, Schwerin, Germany). Sensor array of this analytical instrument is composed of ten different MOS positioned in a small chamber. A description of the ten metal-oxide semiconductors has been given in our previous works (Hong et al., 2012). There are two kinds of data obtained from the e-nose, one is R (the resistance value of the sensors when the sample gas flow through them), the other is G/G_0, where G and G_0 are the conductivities of the sensor when exposed to the sample gas and the zero gas, respectively. The R value may be affected by environmental conditions (such as temperature and humidity) or a day-to-day sensor drift. While on the other hand, the conductivities ratio G/G_0 could reduce the influence of these problems because every responding signal would be calibrated by the responding of sensor when contacting the zero gas. The G/G_0 value is more reliable, thus, it was chosen as the e-nose responding signal.

Taste analysis was performed with an α-Astree e-tongue (Alpha MOS company, France). This taste sensor consists of an array of seven liquid cross-sensitive electrodes or sensors (ZZ, BA, BB, CA, GA, HA and JB), a 16-position auto-sampler and associated interface electronic module. The sensitivity of the seven chemical sensors is different from that of the five tastes (Sourness, Saltiness, Sweetness, Bitterness, and Savoury). Specific description of the sensors and their attributes has also been given in our previous works (Wei et al., 2009).The sensors are made from silicon transistors with organic coatings that govern the sensitivity and selectivity of each individual sensor. The potentiometric difference between each individually coated sensor and Ag/AgCl reference electrode in the equilibrium state was measured and recorded at room temperature.

2.3. Experimental procedures

2.3.1. E-nose sampling procedure

The problems of sensor drift and humidity during e-nose measurement were taken into consideration. As for the sensor drift problem, it was taken care of by controlling the environment parameters (temperature and humidity) and by choosing calibrated data as the initial data. As for the (sample) humidity problem, in contrast to directly e-nose measurement, a pretreatment of employing anhydrous sodium carbonate as desiccant was conducted to observe if reducing of water vapor would improve the performance of e-nose. 100 samples (25 replications $\times$ 4 adulteration levels) were prepared for directly e-nose measurement and e-nose measurement with a pretreatment, respectively. For directly e-nose measurement, each sample (10 mL of cherry tomato juice) was placed in a 500 mL airtight glass vial that was sealed with plastic wrap. The glass vial was closed for 10 min (headspace-generation time) while the headspace collected the volatiles from the samples. For e-nose measurement with a pretreatment, an addition of 5 g of anhydrous sodium carbonate was placed on a filter paper that was placed 4 cm above the bottom of the glass vial. The rest procedure was the same as the directly e-nose measurement. During measuring process, the headspace gaseous compounds were pumped into the sensor arrays through Teflon tubing that was connected to a needle, causing the ratio of conductance of each sensor changed. The measurement phase lasted for 70 s, which was long enough for the sensors to reach stable signal values. Signal data from the sensors were collected by the computer once per second during the measurements. When the measurement process was complete, the acquired data were stored for later use. After each experiment, calibration procedure was carried out to reduce the influence of external parameters such as variation in the relative humidity of the air, changes in the temperatures and the drift of the sensors over time, using zero gas (air filtered by active carbon).

2.3.2. E-tongue sampling procedure

100 samples (25 replications $\times$ 4 adulteration levels) were prepared for e-tongue detection. During the experiment, 80 ml of each cherry tomato juice sample was injected into a 120 ml beaker for e-tongue detection. The measuring time was set to 120 s for each sample, and the sensors were rinsed for 10 s using ultra-pure water to reach stable potential readings before detecting the next sample.