

## VISION SYSTEM AND ELECTRONIC TONGUE APPLICATION TO DETECT COFFEE ADULTERATION WITH BARLEY

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Time consuming and expensive methods have been applied for detection of coffee adulteration based on the literature. In the present work, an optical method (vision system) and the application of an electronic tongue is proposed to reveal the addition of barley in different proportion to coffee in ground and brewed forms. In a range of 1 to 80% (w/w) Robusta coffee was blended with roasted barley. Principal Component Analysis (PCA) accomplished on vision system image data showed a good discrimination of the adulterated samples. The results of Polar Qualification System (PQS) data reduction method revealed even small differences in the right barley content order by point method approach. With Partial Least Squares (PLS) regression the amount of barley in Robusta was predicted with high  $R^2$  (0.996) and relatively low RMSEP (~2%) values in case of vision system data processing. Considering electronic tongue measurements, PCA results showed a good discrimination of samples with higher barley concentration. Misclassification was found in the low concentrated area by Linear Discriminant Analysis (LDA). To obtain an accurate model for barley content prediction in coffee, the most sensitive sensor signals were used to apply PLS regression successfully ( $R^2=0.97$ , RMSEP=3.99% (w/w)).

**Keywords:** adulteration, coffee, vision system, electronic tongue

Mainly two varieties of coffee are used for commercial purpose, known as *Coffea canephora* L. (Robusta) and *Coffea arabica* L. (Arabica). Due to finest organoleptic features, Arabica coffee is mostly preferred by consumers and thus higher prices are obtained. The great difference in the final sale price depends on factors including coffee species (20–25%) and geographic origin. The vulnerability to adulteration with regard to substitutions by several biological materials requires assurance of quality (PIZARRO et al., 2007). These materials are usually agricultural residues, including twigs, coffee husks, parchment, spent coffee grounds, and also other roasted grains, such as corn, barley, maize and soybean (SINGHAL et al., 1997). Nevertheless there are some countries where cereal–coffee mix is consumed, but if the mixing ratio is mislabelled the consumers are mislead. Several papers deal with detection of frauds in coffee mostly based on analytical methods. In case of Arabica coffee, the detection of fraudulent corn was performed by HPLC based on tocopherol determination (JHAM et al., 2007). OLIVEIRA and co-workers (2009) successfully used SPME-GC-MS to detect as low as 1% (w/w) roasted barley in roasted coffee samples, for the darkest degrees of roast. In the

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work of HERNÁNDEZ and co-workers (2008) a vision system was applied to describe the brightness and surface kinetics of Colombian Arabica coffees roasted at different temperatures. Online image analysis made a comparison possible between the measured grey level and a target grey level at each time point. The obtained roasted coffee after this experiment was very similar to the target coffee, and the prepared coffee brew was accepted by the laboratory team. The quantification of adulteration was realized in roasted coffee powder by digital image processing in the study of SANO and co-workers (2003). Image processing of pictures prepared on Arabica coffee mixed with coffee husks and straw, maize, brown sugar, and soybean was realized by grey-scale intensity scale. With this method, good correlation was achieved ( $r=0.90\text{--}0.99$ ) to quantify the amount of adulteration depending on different types of coffee substitutes. New analytical approaches employing cross selective sensors simultaneously, known as Electronic tongues (ET) or taste sensors, can provide important information to extend the application of the device. Considering the authentication and thus especially the analysis of coffee as well as the usage of potentiometric sensors, most of the analysis are accomplished to classify different beverages (LEGIN et al., 1997) or to obtain sensory profiles of coffee samples (TOKO, 1998). The possibility to detect adulteration by electronic tongue was tested on wine (PARRA et al., 2006; STÓJ, 2011) and on cherry tomato juices (HONG et al., 2014).

However, a lack of analysis to confirm authentication of coffee is observed using ISFET sensors although the ability to discriminate coffee blends with different Arabica–Robusta ratios was tested successfully (VÁRVÖLGYI et al., 2012; SOÓS et al., 2013). Hence, the objective of this investigation is to establish an optical method and to extend the application of the electronic tongue for the detection of fraudulent barley in coffee.

## 1. Materials and methods

### 1.1. Materials

Mixtures from different concentrations of 100% Robusta and 100% barley were prepared for the measurements. Six coffee samples were created as mixtures from 1 to 80% (w/w) of barley (B) in Robusta (R): 99R1B, 95R5B, 90R10B, 80R20B, 50R50B, and 20R80B. Two commercially available cereal coffee samples with known Robusta–barley ratios, Ot (51R49B) and AF (47R53B), were provided by the same coffee factory as the pure samples. 100R indicates the analyzed pure Robusta coffee.

### 1.2. Measurement by vision system

The applied vision system was composed of a Hitachi HV-C20 3CCD with Canon TV Zoom lens. Image size was  $768 \times 576$  pixel and the optical resolution was 0.0833mm/pixel. The diffuse light was provided by special geometrical settings of 12 halogen lamps (20W, colour temperature: 3200 K) for the experiment. 100R, 99R1B, 95R5B, 90R10B, 80R20B, 50R50B, 20R80B, AF, and Ot ground roasted coffee samples were measured by vision system. There was no special sample preparation. A circular metal sample holder, opened at the top, was used for the measurement. The same quantity and uniform distribution of the sample was provided with the help of a ruler. After taking the picture, the sample was put back in its container and was mixed with the rest of the same sample for the next measurement. Twenty repetitions of each sample were performed.

Images were stored in bitmap format (24 bit/pixel). RGB colour parameters of each pixel of the image made by the camera were transformed into the HSI colour system using the equations of GONZALEZ and WOODS (1992). HUE represents the angular location (0 to 360°) of the colour in this system. Saturation values were collected into a histogram according to the HUE angle (sum of saturation). Black and white components were not included in the evaluation, because they do not contain colour, in this way there is no need for segmentation algorithms contrary to other image processing techniques. Only those colour components (HUE values) were selected for image data evaluation that characterize coffee. In that case the sum of saturation was higher than 0.

### *1.3. Electronic tongue measurements*

Alpha ASTREE II (Alpha MOS, Toulouse, France) potentiometric electronic tongue was applied to analyze coffee samples. The ET consists of an automatic sampler unit containing 16 slots for samples. The used potentiometric sensors were based on ChemFET, more precisely ISFET, chemical sensors. All measurements were performed with a specific sensor-array (that includes SRS, GPS, STS, UMS, SPS, SWS, and BRS sensors according to the manufacturer). The detailed description of the instrument was presented in several previous publications (KOVÁCS et al., 2010).

For the measurements 100R, 99R1B, 95R5B, 90R10B, 80R20B, 50R50B, 20R80B, AF, and Ot samples were used. Coffee samples were prepared from 6 g of a given coffee–barley mixture that was poured with 100 ml boiling distilled water. After a filtration process, coffee brews were ten-fold diluted by distilled water. The measurements were performed with the same conditions as in the work of VÁRVÖLGYI and co-workers (2012). Nine parallel measurements of each sample were performed. Calibration of the instrument was performed on a mixture of diluted 100R and 20R80B samples. During the measurement, distilled water was used between the coffee samples for cleaning. Cleaning liquids were exchanged after each three cleaning stages to decrease the possibility of contamination.

### *1.4. Statistical analysis*

The statistical evaluation of vision system data was performed using Polar Qualification System (PQS) method. PQS is a general and powerful data reduction method rooted in the evaluation of NIR spectra. The quality of any spectra like data set is defined as the centre of its polar spectrum (polar coordinate system, where radius is the function of spectral value and angle is a function of wavelength). To compute coordinates of the quality point, there are 3 approaches: the point, line, and surface methods (KAFFKA & SEREGÉLY, 2002). In our research, all mentioned approaches were applied for data evaluation. The radius of the quality point was the function of sum of saturation and the angle was a function of HUE value in our case. The best results were obtained with the point method when the x, y coordinates of the quality point of each coffee–barley sample were calculated.

The steady state of electronic tongue sensor signals was applied as variable for the statistical evaluation, considering an average value calculated from the last 10 seconds of the 120 s recorded signal of each measurement result.

Multivariate analysis is a frequently used tool in the interpretation of the data set, especially if the data have high dimensionality. Principal Components Analysis (PCA), Linear Discriminant Analysis (LDA), and Partial Least Squares (PLS) regression (RICHARDS et al., 2002) were employed as data analysis tools in this study. In case of LDA, three-fold

cross-validation was performed, two-thirds of the samples were used for calibration and one third for validation. The training and the test set were selected to make it possible that each sample contributes at least once in the model building and in the validation as well.

PLS regression models were applied to predict the amount of barley in Robusta–barley mixture according to vision system and to electronic tongue measurement results. PLS regression validation was realized by leave-one-out (LOO) method. The obtained models were evaluated based on their determination coefficient ( $R^2$ ) and Root Means Squared Error of Prediction (RMSEP) (HILL & LEWICKI, 2007). All computations were carried out using the program R 3.0.2 (R Foundation for Statistical Computing, Vienna, Austria). HSI and PQS data were calculated by R-project script developed for that purpose.

## 2. Results and discussions

### 2.1. Results of vision system measurements

*2.1.1. PCA and PQS results accomplished on image data of adulterated coffee samples.* Results of PCA performed on vision system data are represented in Figure 1A. The first two principal components (PCs) contained more than 97% of data variance. In low barley content area the sample groups are overlapping, especially 100R and 99R1B. The other samples having low barley concentration (95R5B, 90R10B, 80R20B) are in the right barley content order along PC1. However, the sample points are very close to each other. In high barley content area the 1% difference is visible by the discrimination of 50R50B and Ot (51R49B) along PC2. Sample groups of AF (47R53B) and 20R80B are close to each other despite of their higher difference in barley concentration. This result was obtained due to possible difference in roasting level of our artificial mixture and commercial product.

A data reduction method was needed due to the complicated handling of HUE spectral data. The point method of PQS was applied to determine the centre of gravity with two coordinates. Therefore, the coffee samples were comparable.

PQS plot of the adulterated coffee samples is shown in Figure 1B. The represented points of coffee samples showed a monotonous change along a curved path in the right order of the barley content in coffee samples. Similar results were obtained as in case of PCA. The commercial samples from the same brands were on the same curve path. In that case the discriminated curved path of commercial samples and that of “artificial” mixtures can be due to their different level of roasting. A larger difference was observed in case of coffee samples containing barley in higher ratio. Even 1% barley content difference showed definitely separated points of the samples (50R50B and Ot-51R49B). In the low concentrated area visible differences were smaller; however the sample groups follow the right barley content order.

*2.1.2. Results of PLS regression for barley content prediction.* Results of PLS model built on vision system data to predict barley content in coffee–barley mixtures are shown in Figure 2. The predicted Robusta/barley content values were correct even in case of the samples having 1% difference between Robusta–barley concentrations. The obtained  $R^2$  value was 0.996 showing close correlation, RMSEP value was relatively low, 1.65 (2.06%). Based on the obtained results, the vision system is a promising tool to detect and to predict barley content in coffee.

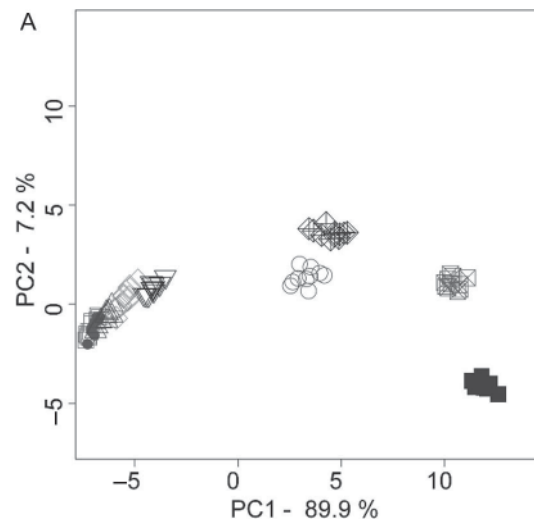


Fig. 1A. PCA of adulterated coffee samples measured by vision system, ■: 20R80B; ○: 50R50B; ▽: 80R20B; ◇: 90R10B; ◆: Ot; ⬢: AF; △: 95R5B; ●: 99R1B; □: 100R

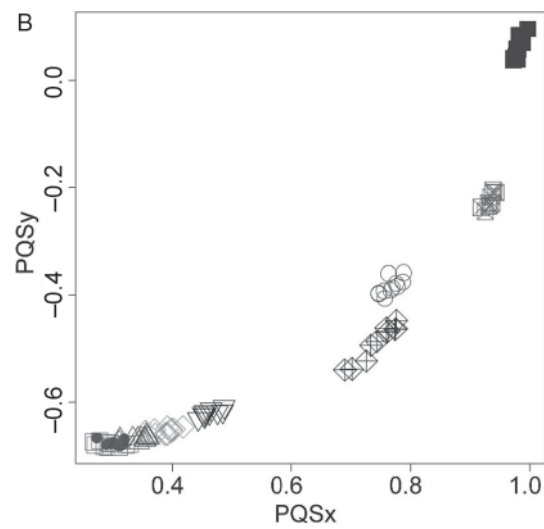


Fig. 1B. PQS plot based on point method of vision system data, for legends see Fig. 1A

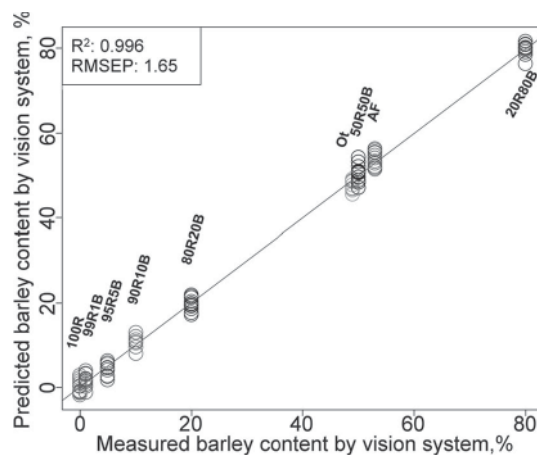


Fig. 2. Prediction of barley content in adulterated coffee by PLS based on vision system image data

## 2.2. Results of electronic tongue measurements

During the preliminary data evaluation performed with PCA on ET data, four outliers for each sample were found. They were excluded from further analysis. Based on showing higher sensitivity and better discrimination ability, five sensors (STS, SRS, SWS, SPS, and UMS) were selected.

**2.2.1. Data exploration and pattern recognition.** PCA plot of different blends of adulterated coffee brews is shown in Figure 3. PC1 (81.5%) and PC2 (15.6%) visualize the information of five sensors in a two-dimensional space containing more than 97% of the data variance. 20R80B, AF (47R53B), Ot (51R49B), and 50R50B samples are well separated. Furthermore, PC1 is a contributing factor, which is related to the concentration (from left to right increasing barley concentration). However, less discrimination capability was observed for the barley concentration in the low concentrated area (0 to 20% w/w), sample groups overlap and they are not in the right concentration order.

PCA accomplished as pattern recognition method only, displays weakness mainly in the low concentrated area. This phenomenon was observed in case of our previous experiments (VÁRVÖLGYI et al., 2012), when the sample points of 100% A (Arabica) and those of 75% A–25% R were close to each other. Hence, LDA is applied to classify adulterated samples.

**2.2.2. Classification by LDA.** Classes for model building were defined according to barley content in coffee. In Figure 4, the discrimination between the classes 100R, 99R1B, and 95R5B (low barley concentration) was not significant, and the sample points of 50R50B and Ot (51R49B) overlapped. The commercial sample AF (47R53B) and 20R80B were discriminated from the other sample groups as well as from each other.

The corresponding LDA-classification matrix (data not shown) showed correct classification (100%). In terms of validation, most of the samples were classified correctly (96.7%), although a sample from Ot-group (49% w/w barley) was assigned to 50R50B (50% w/w barley) in case of electronic tongue measurements.

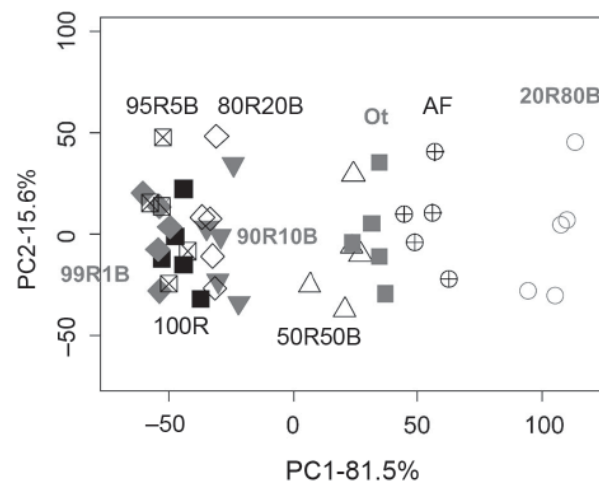


Fig. 3. PCA plot of coffee blends with the results of five sensors of electronic tongue  
 ○: 20R80B; △: 50R50B; ⊕: AF; ■: Ot; ◇: 80R20B; ▼: 90R10B; ⊠: 95R5B; ◆: 99R1B; ■: 100R

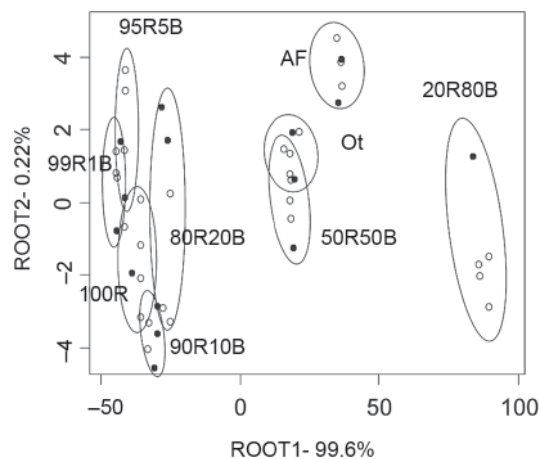


Fig. 4. LDA classification plot of adulterated coffee samples measured by electronic tongue. Empty dots are contributed to calibration set; filled dots to the validation set

**2.2.3. PLS regression model to predict the barley concentration.** In order to predict the barley concentration in coffee, multivariate calibration was applied for electronic tongue technique. Figure 5 displays the prediction model finally selected as optimal with regard to predictive ability.

Despite a high correlation ( $R^2=0.97$ ), the accuracy of prediction was not given for new values based on the sensor signals, especially for 100R or 80R20B. According to 100R, all predicted values exceeded the measured value, whereas all values for 80R20B were predicted below the measured. Thus, no models were acquired with a low RMSEP. Concerning this model, a RMSEP of 4.99 features an accuracy of 3.99 % w/w barley, so that no low level of barley in coffee can be predicted correctly applying these five sensors of ET.



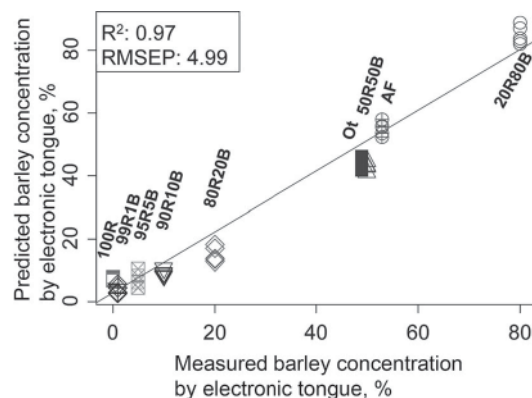


Fig. 5. PLS- regression model to predict barley concentration in coffee blends according to electronic tongue data

### 3. Conclusions

Results of vision system data were promising; PCA showed already a relatively good discrimination in the right barley content order. Sample points on PQS plots described a monotonous change along a curved path following the right barley–coffee ratio. PLS regression model was found suitable to predict the barley content in barley–coffee mixture in roasted-ground form with close correlation ( $R^2=0.996$ ) and low prediction error ( $\sim 2\%$ ). Compared to the work of OLIVEIRA and co-workers (2009), similar detection power of roasted barley in coffee was achieved by vision system method as by SPME-GC-MS. However, in our experiment there was no need for any time consuming sample preparation or the application of expensive analytical instruments.

A good separation with regard to the high concentrated barley level indicates the capability to detect adulteration in coffee by electronic tongue measurement. For the low concentrated level, overlapping of the different samples of the same group is an indication of less sensitivity. Consequently further analyses are proposed with samples having more different Robusta–barley composition in low concentrated area. According to the prediction power of the PLS model, the obtained accuracy was lower than in case of vision system data.

Nevertheless, vision system seems to be an adequate tool to reveal coffee adulteration with barley in ground roasted form and also to avoid mislabelling. Therefore, a further aim is to develop a portable, low cost construction of the vision system to help the work of the authorities and the industry as well. Beside that, electronic tongue systems were considered also in future applications to quantify different adulterants in coffee in brewed form and to gain further information, e.g. organoleptic characteristics.

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