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Theses of doctoral (Ph.D.) dissertation

Application of electronic tongue to estimate quality parameters of wines

János Soós

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Name: PhD School of Food Science

Field: Food Science

Head: **Livia Simon-Sarkadi, DSc**
Department of Food Chemistry and Nutrient
Faculty of Food Science
Szent István University

Supervisors: **Prof. András Fekete †, DSc**
Department of Physics and Control
Faculty of Food Science
Szent István University

Zoltán Kovács, Ph.D
Department of Physics and Control
Faculty of Food Science
Szent István University

Habil. Ildikó Magyar, Ph.D
Department of Oenology
Faculty of Horticultural Science
Szent István University

The applicant met the requirement of the PhD regulations of the Szent István University of Budapest and the thesis is accepted for the defence process.

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approval of Head of School approval of Supervisor

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approval of Supervisor

1. Introduction

Viticulture and wine production still play significant roles in Hungarian culture. Domestic wine producers and wine communities seek to ensure that, all wine qualities such as the region, specific characteristics are all protected in order to uphold the brand and territorial reputation of the wines against wine fraud. In the European Union (EU) there are strict laws for origin protection of wines. All wine regions therefore have their own regulations with mandatory compliance for viticulturist and wine makers. Higher quality wines can be sold at higher prices, there are people who want to exploit this by producing higher quality wine products in a disallowed method. Wine producers must comply with the rules laid down by national and international regulatory authorities. However, in order to reduce costs, irregular treatments such as dilution with water, addition of prohibited substances, use of grapes from different regions or use of prohibited aging methods may occur. Detecting the wine fraud is a very lengthy process often requiring a lot of sensory and analytical measurements until the authenticity of the wine can be determined. Electronic tongue can be an effective tool for control organizations to detect fraud by some producers who, try to mislead consumers by giving incorrect information about the type of grape and its geographical origin. The electronic tongue can be used for quality control of wines and also, by official organizations to verify the authenticity of the product by complementing the results obtained from traditional analytical methods.

Numerous studies have been presented to examine food by electronic tongue, but according to literature, comparisons of ISFET-based sensors have not yet been conducted for wines. The influence of electronic tongue sequence on experimental results have already been demonstrated, but not the effect of the repeat order. Furthermore, numerous studies have shown that you can achieve fast, reliable and reproducible results using electronic tongue. There has been no studies on the distinction between white and red wines from different wine regions of Hungary according to their origin or variety, particularly, with respect to Tokaj wine specialties. In addition to the typical growing areas which can be recognized in wine flavours what may can be estimated by electronic tongue not previously been studied yet. It is therefore necessary to test the applicability of the electronic tongue in measuring different quality parameters in some assorted Hungarian wines.

2. Research aims

The aim of this doctoral dissertation was: to develop an electronic tongue method that permit the detection of the chemical substances present in wine by estimating their quality parameters and basic organoleptic properties. The study focused on two major objectives, each of which is divided into two further sub-objectives.

I. Development and application of a measurement method based on the use of electronic tongue in commercial wine samples to distinguish wines by variety and by region. In addition, exploration of correlations between the results of sensory evaluation, analytical tests and electronic tongue analysis.
   1. Analysis of red wines (4 varieties x 7 wine regions)
   2. Analysis of white wines (4 varieties x 4 wine regions)

II. Applicability of electronic tongue for characterization of Tokaj botrytis wine specialties.
   1. Separation of various *aszú* wines with different proportions of botrytised berries in the light of the new regulation in Tokaj
   2. Distinguish between *aszú, fordítás, szamorodni* and adulterated wines

An essential part of the tasks in the above topics was the development of targeted applications of electronic tongue for wine quality.

Goals of the research were to study:

- The effect of the measurement order of each sample on the electronic tongue results.
- The effect of the dilution ratio of the samples on electronic tongue results.
- Analysis of the correlation between the sensory evaluation and the results of electronic tongue measurements.
- Estimation of the results of sensory evaluation based on the results obtained in electronic tongue.
- Estimation of the chemical parameters of the main types of wine based on the measurement results of the electronic tongue.

3. Materials and methods

3.1 Materials

Initial stages of the research focused on 2012 red wines (Table 1) by comparing results of basic analytical and organoleptic assessment and the data measured by electronic tongue. This was to access the effect of repetition order on electronic tongue measurements.

Table 1. Varieties and regions of the 2012 vintage red wine samples

	Variety	Wine region	Sample code
1.	Cuvée	Eger	EgCu
2.	Cuvée	Kunság	KuCu
3.	Cuvée	Villány	ViCu
4.	Kékfrankos	Ászár-Neszmély	ÁNKf
5.	Kékfrankos	Sopron	SoKf
6.	Kékfrankos	Szekszárd	SzKf
7.	Merlot	Balatonfüred-Csopak	BCMe
8.	Merlot	Eger	EgMe
9.	Merlot	Szekszárd	SzMe
10.	Portugieser	Eger	EgPo
11.	Portugieser	Szekszárd	SzPo
12.	Portugieser	Villány	ViPo

Taking into account the results obtained, white wines from 2013 (Table 2) were also analysed.This was compared the results of two sensor arrays of electronic tongue with the results of analytical and sensory assessment.

Table 2. Varieties and regions of the 2013 vintage white wine samples

	Variety	Wine region	Sample code
1.	Chardonnay	Balatonboglár	BbCh
2.	Chardonnay	Etyek-Buda	EBCh
3.	Chardonnay	Mátra	MaCh
4.	Chardonnay	Villány	ViCh
5.	Cserszegi fűszeres	Balatonboglár	BbCs
6.	Cserszegi fűszeres	Etyek-Buda	EBCs
7.	Cserszegi fűszeres	Mátra	MaCs
8.	Cserszegi fűszeres	Villány	ViCs
9.	Sauvignon blanc	Balatonboglár	BbSb
10.	Sauvignon blanc	Etyek-Buda	EBSb
11.	Sauvignon blanc	Mátra	MaSb
12.	Sauvignon blanc	Villány	ViSb
13.	Pinot gris	Balatonboglár	BbSz
14.	Pinot gris	Etyek-Buda	EBSz
15.	Pinot gris	Mátra	MaSz
16.	Pinot gris	Villány	ViSz

In the second part of the studies the 2014 vintage of self-made *aszú* samples of the changing light of the specification were analysed (Table 3). The lower quality *aszú* no longer meet the new requirements. In this case, in order to investigate the higher content of Aszú, the efficiency of three different dilutions was examined to obtain the optimal dilution ratio used for electronic tongue measurements.

Table 3. Tested *aszú* samples according to the updated Tokaj wine region regulations

	Name of wine	Noble rotten berries/ extracting wine	Wine regulation of Tokaj	Sample code
1.	Furmint base wine	-	-	Alap1
2.	*Aszú* 3p	0.6kg/L	Pre-2013	Aszu3p
3.	*Aszú* 4p	0.8kg/L	Pre-2013	Aszu4p
4.	*Aszú* 5p	1kg/L	Post-2013	Aszu5pN
5.	*Aszú* 6p	1.2kg/L	Post-2013	Aszu6pN
6.	*Aszú* Essencia	1.4kg/L	Post-2013	Aszu7pN
7.	*Aszú* 5p1	-	Blend	Aszu5p1
8.	*Aszú* 5p2	-	Blend	Aszu5p2

Finally, self-made *aszú* and *fordítás* samples from 2016 were studied to detect changes in the quality according to the maceration time of noble rotten berries (Table 4). Therefore, comparative studies with commercially available *aszú* and *szamorodni* samples was conducted with artificially adulterated wine. Wine fraud in this case reflects a very extreme case of not using *aszú* berries for the wine production. These adulterated wines are very different analytically and organoleptically from wine specialties, so in reality there is no such adulteration. In practice, partial replacement of aszú berries with must concentrate may be more likely, so series of adulterations were also investigated by setting the wines to the same sugar content with different proportions of *aszú*, base wine and must concentrate (Table 5).

Table 4. Preparation of adulterated wines and *aszú, fordítás* samples based on the quality of noble rotten berries (I, II) and maceration time (24, 48 hours)

	Name of wine	*Aszú* berries quality	Maceration time (h)	Sample code
1.	Furmint basewine	-	-	Alap2
2.	Basewine+sugar (Adulteration)	-	-	Alap_cuk
3.	Basewine+concentrate (Adulteration)	-	-	Alap_sur
4.	*Aszú* I/1	I	24	Aszu_I_1
5.	*Aszú* I/2	I	48	Aszu_I_2
6.	*Aszú* II/1	II	24	Aszu_II_1
7.	*Aszú* II/2	II	48	Aszu_II_2
8.	*Aszú* blend 1	I és II (1:1)	24	Aszu_Vegy1
9.	*Aszú* blend 2	I és II (1:1)	48	Aszu_Vegy2
10.	*Fordítás* I/1	I	24	Ford_I_1
11.	*Fordítás* I/2	I	24	Ford_I_2
12.	*Fordítás* II/1	II	24	Ford_II_1
13.	*Fordítás* II/2	II	24	Ford_II_2
14.	*Fordítás* blend 1	I és II	24	Ford_Vegy1
15.	*Fordítás* blend 2	I és II	24	Ford_Vegy2
16.	*Aszú* (Furmint)	NA	NA	Aszu_Furm
17.	*Aszú* (Zéta)	NA	NA	Aszu_Zeta
18.	*Szamorodni* (Hárslevelű)	-	-	Szam_Hárs
19.	*Szamorodni* (Zéta)	-	-	Szam_Zeta

Table 5. Adulterated *aszú* series with base wine and must concentrate

Sample code	*Aszú*/basewine ratio		Aszu_Vegy2 (ml)	Alap2 (ml)	Concentrate (ml)	Sugar contetnt (g/l)
Aszu100	100	0	300.00	0.00	0.00	178.10
Aszu99	99	1	297.00	3.00	0.51	178.10
Aszu95	95	5	285.00	15.00	2.57	178.10
Aszu90	90	10	270.00	30.00	5.13	178.10
Aszu80	80	20	240.00	60.00	10.27	178.10
Aszu70	70	30	210.00	90.00	15.40	178.10
Aszu60	60	40	180.00	120.00	20.53	178.10
Aszu50	50	50	150.00	150.00	25.66	178.10

3.2 Methods

Electronic tongue

The study involved the Alpha Astree 2 electronic tongue, which is a potentiometric electronic tongue. During the red wine measurements, the effect of the repeat was examined on the measurement results of the electronic tongue by using three measurement methods (full sequence repetition, partial sequence repetition, repetition per sample). In the case of white wine measurements, two types of sensor arrays were used and compared under the same conditions. One of the sensor arrays recommended for measuring food was ZZ, BB, CA, GA, HA, JB. The other was the specific sensor array, which was used to measure red wines too. Some of the sensors according to the manufacturer, are somewhat specific to certain base flavours SRS - sour; BRS - bitter; UMS - umami; STS - salted; SWS – sweet. For white and red wines, 50% dilution was used because of the potential masking effect on the sensors due to the higher sugar content. During the wine specialities experiments, preliminary studies had to be performed to find out which degree of dilution was best suited for determination of the *aszú* samples. The samples were measured without dilution and with distilled water (samples contain *aszú* in 50% and in 25%), in nine replicates. Data was processed using principal component, discriminant analysis, and partial least squares regression (three-fold cross-validation), after excluding outliers and using drift correction.

Analytical methods

The routine analyses were carried out in the Research Laboratory of the Department of Oenology of the SZIU, in accordance with the general wine analytical practice according to analytical methods adapted by the International Organisation of Vine and Wine (OIV). The titratable acidity, pH, sugar, alcohol and volatile acid content of all red and white wines were examined. Previous measurements were supplemented with the determination of total polyphenol, catechin, leucoanthocyanin, glycerol and gluconic acid contents during the *aszú* measurements.

Sensory evaluation

In the case of white and red wine tests, for the sensory evaluation the 12 wine samples were divided into two groups and two wine samples were used as overlapping wine in both groups so seven wine samples were evaluated in each group. The sensory evaluation was carried out by 12 panellists according to the profile analysis (MSZ ISO 11035:2001). The test involved an unstructured scale, where the panellists had to compare the wine samples according to the given criteria. As a first step in evaluating the intensity values indicated by the panellists, the results were converted to a percentage. Processing of the results were done with a selection of reviewers, then one-way analysis of variance (ANOVA) and Tukey-HSD test.

4. Results

4.1 Analysis of red and cuvée wines

Full sequence repetition

In order to maximize the differences between red wine groups, discriminant analysis was performed. The separation of the samples is shown in Figure 1, which showed that the ANKf group was most different from the other analysed wine samples. On the basis of the discriminant analysis it can be stated that the electronic tongue always distinctively grouped the samples resulting in a correct classification rate of 100%, but during the three-fold cross validation one sample was misclassified (the EgCu with the EgPo) so the error of the validation was 0.93%.

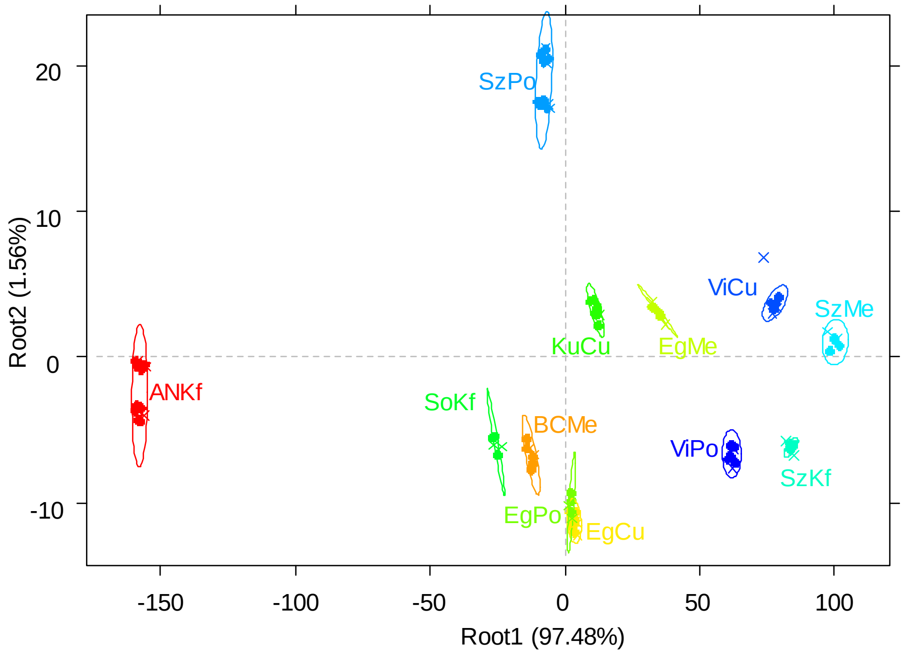

Figure 1. LDA (Root1-2, solid circle calibration (2/3), x-validation (1/3), ellipse-95% confidence interval) Red wine discrimination (full sequence repetition)

Partial sequence repetition

The results of the partial sequence repetition (the samples were measured in 3 repeat) showed that this measurement method is less suitable for distinguishing red wines. Based on the cross-validation matrix of the red wine samples, only the ANKf and SzKf samples were not misclassified, the accuracy of the calibration model was 74.58%. Overall, the classification model was able to classify with an accuracy of 60.15%, which is far below the accuracy of the previous method.

Based on these results it can be concluded that the partial sequence repetition is not capable of distinguishing the red wines. This is probably due to the fact that the signals belonging to a sample can be divided into three groups according to their intensity and the drift correction applied did not allow these differences within the sample to be sufficiently reduced or even disappear.

Repetition per sample

The discriminant analysis showed that this method can also effectively separate red wine samples from each other (Figure 2). In the case of cross-validation the results showed that the model building proved to be 100% correct. During the validation of the model 0.93% misclassification was observed, just as in the case of full sequence repeats, except that in this case SzKf group was misclassified as ViCu group.

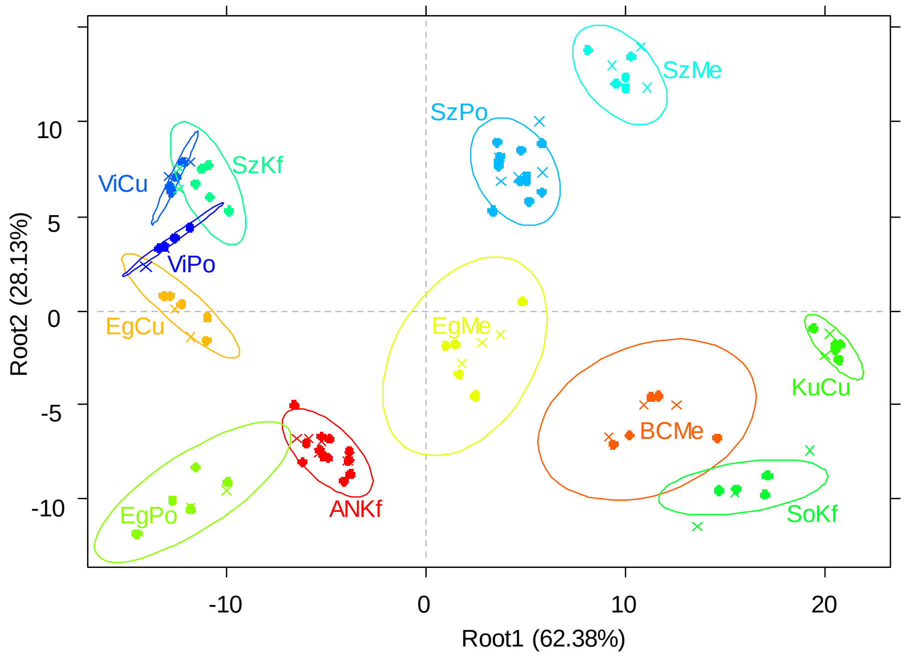

Figure 2. LDA (Root1-2, solid circle calibration (2/3), x-validation (1/3), ellipse-95% confidence interval) Distinguishing red wines from electronic tongue data (repetition per sample)

Prediction of the chemical parameters showed that the closest correlations can be reached with the full sequence repetition method (e.g.: full sequence repetition: R^2_{acid}=0.95; partial sequence repetition: R^2_{acid}=0.78; repetition per sample: R^2_{acid}=0.82). The highest coefficient of determination was obtained for the prediction of pH, which was higher than 0.98 in the validation. The method can be used by wineries to estimate the most important chemical parameters of red wines without the use of chemicals. All in all, the first measurement method proved to be the most effective for the separation of red wines and for the estimation of the organoleptic and chemical parameters, so this method was used in further experiments.

4.2 Analysis of white wines

Electronic tongue measurements were performed with two sensor arrays based on the first measurement method described in the previous chapter. In this case the samples were measured in a single sequence. Drift correction and outlier detection was carried out on the obtained data.

Results of sensor array for food

The 16 samples tested could be successfully separated with 100% accuracy during the calibration and the validation error was only 3.06% according to the LDA. The Sauvignon blanc from Balaton showed the greatest separation along Root1, which describes 83.12% of the difference (Figure 3). EBCs and MaSz samples have the lowest acid content, so it can be stated that in this case Root1 showed a patten of increasing acidity.

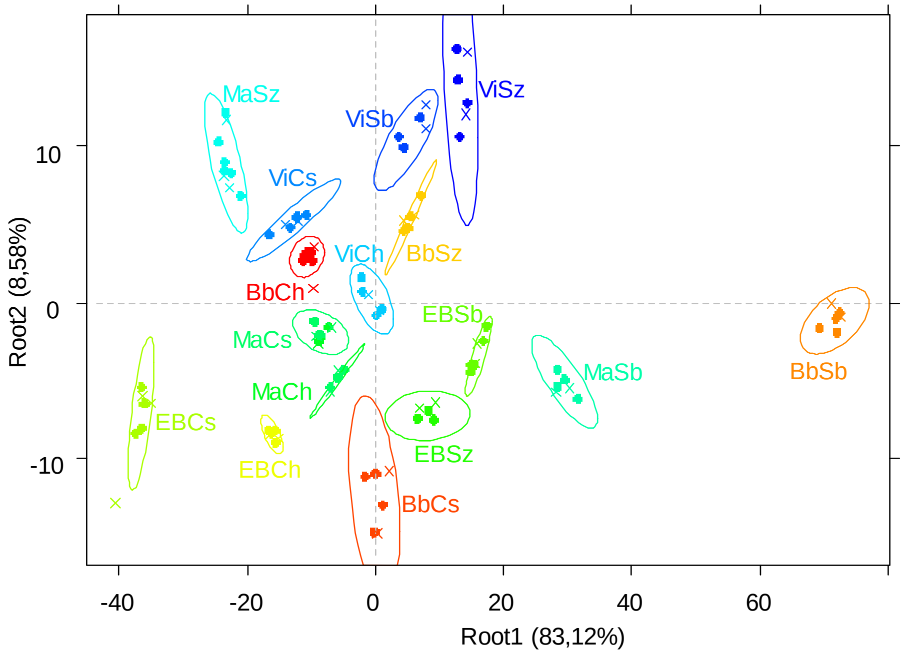

Figure 3. LDA (Root1-2, solid circle calibration (2/3), x-validation (1/3), ellipse-95% confidence interval) White wine discrimination from electronic tongue data

Analysing all wine from four wine regions, there was an overlap between the wines from the Etyek and the Mátra wine region, which can be recognized in calibration (7.69%) and validation (12.12%) errors. Only these two groups could not be separated based on the data of the electronic tongue. When the wines from Etyek-Buda were excluded from the analysis, there were fewer misclassifications during calibration and validation, thus reducing the error values to 1.37% and 2.74%, respectively.

The varieties were also examined by discriminant analysis. The results showed that the overlap between the Cserszegi Fűszeres and Chardonnay samples was so large along Root1 and Root2 that the calibration error was 26.03% and that of the validation was 28.77%. These errors were significantly reduced when Chardonnay samples were excluded from analysis. In this case the correct classification was 99.07% during the calibration and 96.33% during the validation.

Results of specific sensor array

The result of the discriminant analysis showed that the electronic tongue was able to classify the different white wine samples with 100% correct accuracy and proved to be 100% correct during the validation as well.

Discriminant analysis was also performed for with the groups being the production areas. Based on the results it can be stated that the specific sensor array could not effectively separate the tested wines by region. The model building showed 53.25% recognition and 38.46% validation accuracies. The results did not improve so much even when similar exclusion was performed like in the previous measurement. Even without the wines of the Etyek-Buda wine region, only 76.74% showed correct classification during the model building.

The next discriminant analysis was carried out for the varieties of the samples. In this case better results were achieved. The electronic tongue was able to successfully discriminate the varieties during model building (88.31%), this value increased to 93.02% without Chardonnay wines, but during the validation of the model the correct classification decreased from 76.92% to 72.73% without the Chardonnay samples.

Generally, it can be stated that both sensor arrays were capable of discrimination white wines. However, thr sensor array recommended for food measurements could effectively separate wine samples by variety and region with better accuracies. Furthermore, the prediction of the sensory attributes and the chemical parameters also showed that a closer relationship can be found with the sensor array recommended for food measurements (e.g.: specific sensor array: R^2_{acid}=0.92, $R^2_{acidic\_taste}$=0.76; sensor array for food measurement: R^2_{acid}=0.94, $R^2_{acidic\_taste}$ =0.90). Based on the results this sensor array was more suitable for the analysis of wines, so this sensor array was used for the further experiments.

4.3 Investigation of *Aszú* wines in the light of the new wine regulation in Tokaj

The results of the discriminant analysis showed that the base wine is very different from the *aszú* samples, subsequent data evaluations were performed without it. Calibration and validation did not result in misclassification at 50% dilution. In addition, the electronic tongue was capable of separating the *Aszú* at 50% dilution according to the old and new regulations, and could clearly classify the mixed *Aszú* created by their blending.

For each chemical parameter, partial least squares (PLS) regressions were performed for all three dilutions. The alcohol, acid, sugar, glycerol, gluconic acid content and the pH value of the samples were well estimated. Of these, the estimation of sugar content is illustrated in Figure 4 for each of the three dilution ratios.

The results showed that the best prediction could be obtained with 50% dilution in most cases but with two exceptions, as in the prediction of acidity and gluconic acid content 25% dilution was showed closer relation. Total acids, sugars, glycerol and gluconic acid were found in significantly higher concentrations in *aszú* wines than in normal wines. Thus, the prediction of these parameters played a key role in the description of *aszú* wines. The volatile acid content of the samples was the least predictable parameter. Generally, 50% dilution was the most ideal for measuring *aszú* and was adapted in the further experiments.

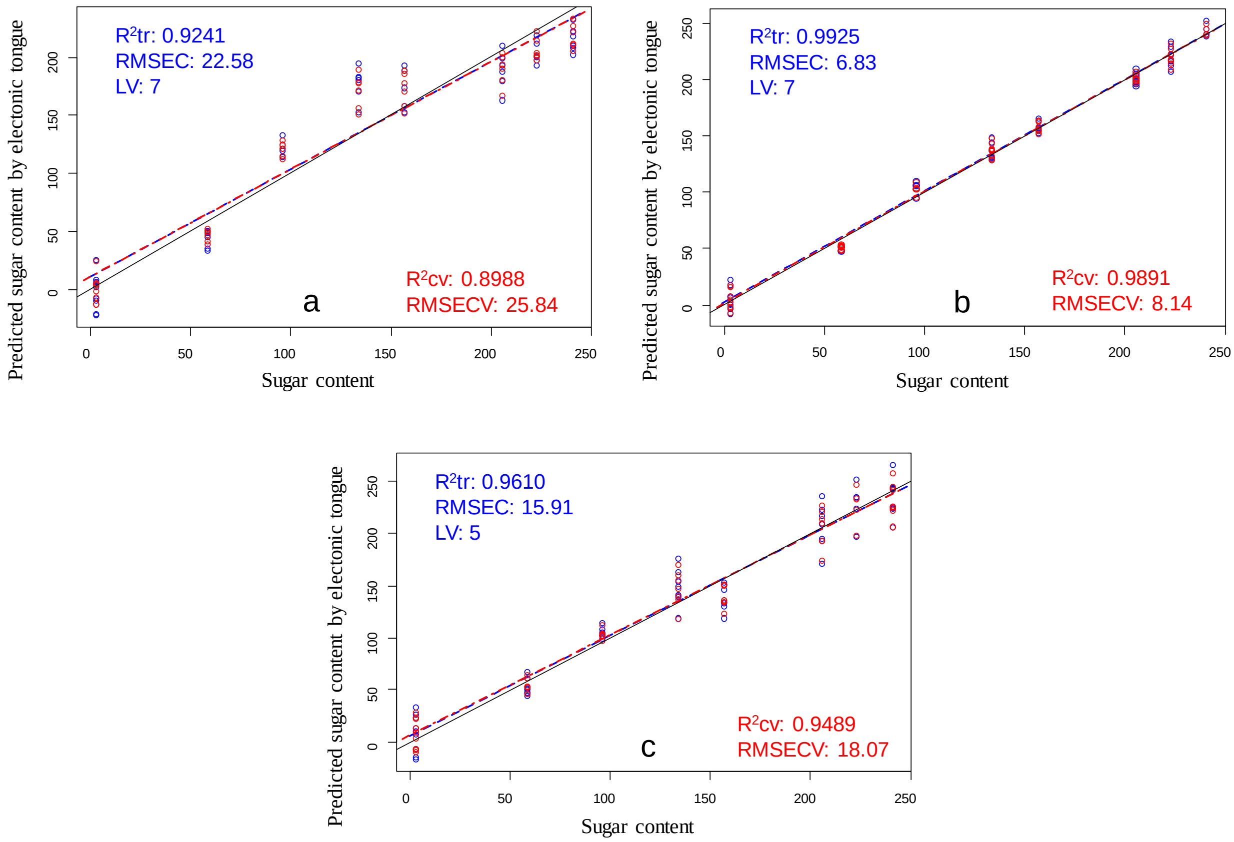

Figure 4. Prediction of sugar content as a function of dilution from electronic tongue data (a, 25%; b, 50%; c, 100%)

4.4 Analysis of *aszú*, *fordítás* and wine fraudulent

Self-made wine specialties samples were compared with commercially available *aszú* and *szamorodni* samples based on analytical parameters and electronic tongue measurement results. The chemical parameters of commercial samples were predicted based on the models developed from the chemical parameters of self-made wine specialties and electronic tongue data. In addition, lowest levels of adulteration were investigated to check the capability of electronic tongue for such discrimination.

During the first the discriminant analysis each sample formed a separate group. Adulterated wine showed the biggest differentiation among the groups. The classification was correct with 100% accuracy during calibration and validation.

For better understanding of the differences between *aszú* and *fordítás*, the discriminant analysis was carried out without adulterated wines (Figure 5). *Aszú* samples produced from first-class *aszú* berries showed the biggest discrimination. The group of Aszu_Vegy1 was close to the group of *aszú* samples made from second-class *aszú* berries along Root1. Commercial *Aszú* wines along Root1 were the most similar to *Aszu_Vegy1*, while *Szam_Hars* aligned with *Ford_Vegy2*. In addition, it could be seen that the *Ford_vegy1* group were close to the *fordítás* sample made from first-class a*szú* berries while *Ford_vegy2* were close to the second-class *fordítás*.

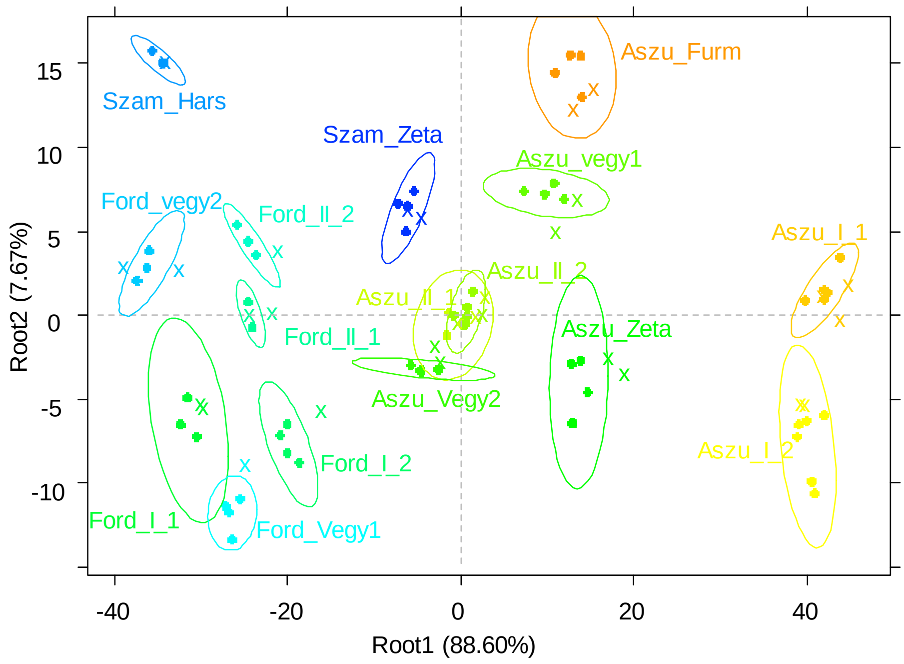

Figure 5. LDA (Root1-2, solid circle calibration (2/3), x-validation (1/3), ellipse-95% confidence interval) Discrimination of *aszú* and *fordítás* and commercial samples based on electronic tongue measurement

In the following discriminant analysis, a categorization was made that included all of the self-made *aszú*, and other groups for all the *fordítás*, commercial *aszú* samples, *szamorodni* samples and adulterated wines samples (Figure 6). The adulterated wines showed the biggest separation along Root1. The other samples showed differences along Root2. The model classification accuracy was 95.77% during model building and 92.75% in model validation.

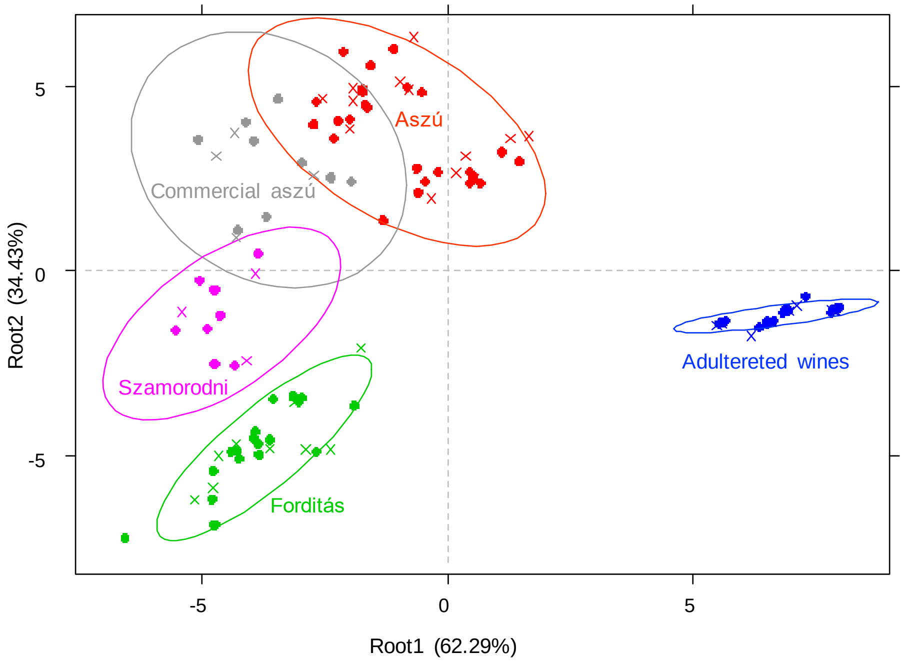

Figure 6. LDA (Root1-2, solid circle calibration (2/3), x-validation (1/3), ellipse-95% confidence interval) Disrimination of aszú, fordítás, adulterated wine samples and commercial samples based on electronic tongue measurement

Based on the results it can be concluded that the electronic tongue was able to discriminate the *aszú* samples from the adulterated wine samples. The *fordítás* samples could also be separated from the *aszú* samples without any error. The commercial aszú samples were the most similar to those *aszú* samples made from the mixture of first and second class *aszú* berries. The same statement could be concluded on the basis of analytical parameters.

For the PLS model building only the self-made *aszú* and *fordítás* samples were used. The obtained models were validated and used for the prediction of the chemical parameters of the commercial samples. The acid, sugar, total polyphenol, catechin, leucoanthocyanin, and glycerol content of the samples were well predictable, as well as the pH value. Prediction of gluconic acid in this case did not show a sufficient close correlation, but the prediction of the glycerol / gluconic acid ratio, which plays a significant role in the quality of *aszú* showed close correlation with the electronic tongue data. In cases where high values of R^2 ($R^2 > 0.9$) were obtained during the validation, the parameters of the commercial *aszú* samples were predicted using the established experimental models.

Figure 7 illustrates the prediction of sugar content. Regression model fitted to the groups of self-made Tokaj wine specialties had a R^2 value of 0.9348 for sugar content, which was a very close correlation.

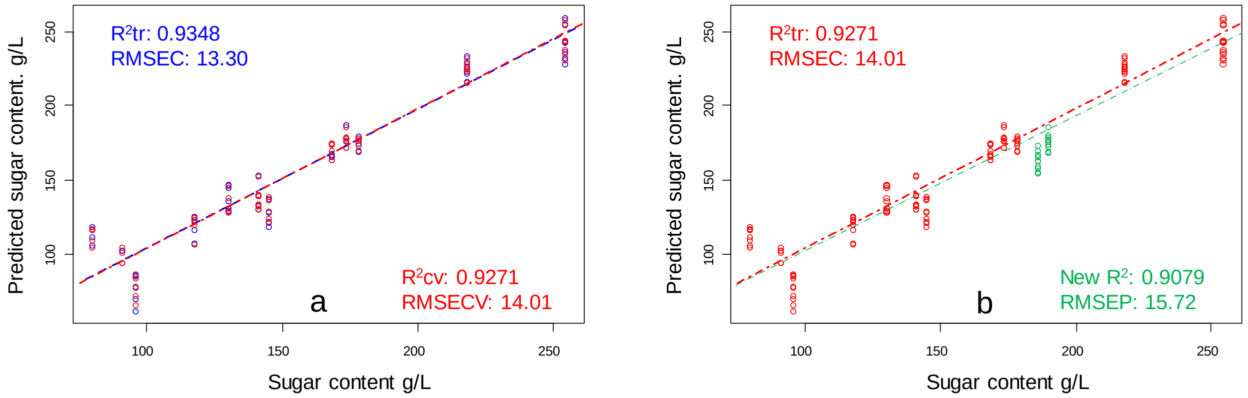

Figure 7. PLS regression for prediction of the sugar content of self-made Tokaj wine specialties (a) and the prediction of sugar content of commercial aszú samples projected into the obtained model (b)

In addition, how to estimate the degree of adulteration compared to the original aszú sample using PLS model (Figure 8) was investigated. The regression model fitted to the groups of the self-made adulterated wine series had a R^2_{CV} value of 0.9856 for validation, the validation error was low RMSECV =2.05. The built model could give a good opportunity to recognize the adulteration if *aszú* sample contains concentrated must in more than 2%.

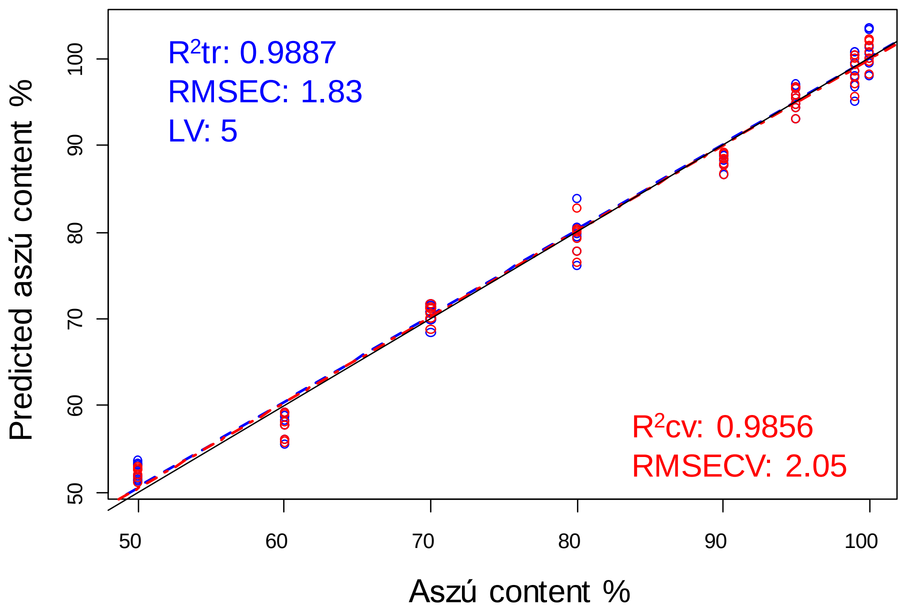

Figure 8. Prediction of the *aszú* content of adulterating level using a PLS model

5. New scientific results (Thesis)

Based on the results of the measurements of the Alpha Astree electronic tongue, the following new scientific contributions were made to literature. These results are fully validated for the instrument / sensor array and samples used in the experiments, but in actually contributes significantly to our knowledge of electronic tongue. The significance of the thesis is increased by the fact that with the help of an electronic tongue instrument the investigations of the possible unauthorized substances (sugar, must concentrate) in Tokaj wine specialties have been ascertained.

1. New measuring method based on electronic tongue was developed to monitor wine samples. It was proven with the evaluation (multivariate method PCA, LDA) of data that the order of repetition greatly influences the separability of the samples to be measured and has a significant influence on the prediction of the sensory attributes and analytical parameters. By analysing red wines, the three different measurement methods (full sequence repetition, partial sequence repetition, sample repetition) was used, the full sequence repetition method showed the best results and reached 100% correct classifications for the wine variety. Among the PLS results of organoleptic properties, sensory sweet and acidic taste intensities showed the closest correlations ($R^2_{sweet\_taste}$=0.8314 and $R^2_{acidic\_taste}$=0.8282) with the electronic tongue data. Among the analytical properties, pH and acid content showed the closest correlations (R^2_{pH}=0.9887 and R^2_{acid}=0.9503).

2. After comparative measurement of the sensor arrays, the sensor array recommended for food measurements could effectively differentiate the white wines by variety and wine region with better accuracies. These included Cserszegi fűszeres, Sauvignon blanc and Pinot gris wines. The measurements proved that the sensory array recommended for food measurements can be more effective in predicting the sensory attributes and analytical properties of white wines than the specific sensor array. Among the predicted sensory attributes from the results of the electronic tongue, the acidic taste intensity showed the closest correlation (R^2=0.9046), and from the analytical parameters it was also the acid content (R^2=0.9432).

3. It was proved that between the 25-50% distilled water dilutions and the undiluted wines, the 50% dilution ratio was ideal for measuring *aszú* wines using the electronic tongue. Among the predicted analytical parameters from the results of the electronic tongue, the sugar and acid contents showed the closest correlations (R^2_{sugar}=0.9891 and R^2_{acid}=0.9752).

4. It was determined that the electronic tongue is suitable for the monotonous monitoring of chemical changes in *aszú* wines made from the different ratios of the used aszú berries. The correct grouping ratio reached 100% accuracy with the 50% dilution ratio used.
5. By measurements it was proved that Tokaj wine specialties made from using two qualities of noble rotten berries can be discriminated on the basis of electronic tongue measurements, both in terms of *aszú* and *fordítás*. The LDA models showed good classification with 100% accuracy.
6. It was verified that the models built using *aszú* sample made under controlled conditions were able to predict the main chemical attributes of commercial samples based on electronic tongue data and showed close correlation between the acid (R^2=0.9014; RMSEP=0.26), sugar (R^2=0.9079; RMSEP=15.72) and glycerol content (R^2=0.9098; RMSEP=1.22).
7. Based on the results obtained with the electronic tongue, the content of the must concentrate of *Aszú* can be predicted with a close correlation (R^2=0.9856, $RMSE_{cv}$=2.06%), thus this kind of adulteration can be detected with electronic tongue.

6. Conclusions and suggestions

This study required significant methodological development, during which several method variables and measurement techniques from the point of view of efficiency (eg repeat order, dilution ratio, sensor type, statistical methods) were taken into account. The results of the research provided the basis for the electronic tongue to be an effective tool for wine analysis. For this instrument to appear in practice, further measurements in the operating environment are required, but the results of this research may predict the use of the instrument.

It could also help winemakers in several areas. Even during the harvest, the quality of the musts at harvesting time must be determined, since it is a common problem to determine the quality of the grape or must, purchased from the grower. Electronic tongue as an objective tool, could produce reliable results that both parties can work with.

Because very small changes can be detected in must or wine, electronic tongue could be used to detect taste diseases in wine before they can be detected by human taste perception, which could be a big help for winemakers. Another exciting area of study for Tokaj wine specialties may be the estimation of the maturation period in wooden barrels or the discrimination between Tokaj sweet wines and other botrytis wines in the world (e.g.: Sauternes, trockenbeerenauslese, etc.). Electronic tongue can be successfully used not only in large-scale wineries but also in smaller family wineries, where it could be used as a continuous monitoring tool to maintain the quality of the wine.

Due to the size of the electronic tongue, it is not suitable for on-site analysis. In further research, it may be worth extending the analysis to another potential adulterant. It could be a great help if a more portable or handy instrument is developed to assist the regulatory authorities in carrying out inspections as a quick method. Nonetheless, could system can be incorporated into electronic tongue functionalities for online applications. Reducing the number of sensors would also reduce the size of the instrument.

7. Publications related to the topic

Articles with impact factor

János Soós, Evelin Várvölgyi, Lajos Dénes Dénes, Zoltán Kovács, József Felföldi (2014) Application of electronic tongue to discriminate white wines originated from different regions of Hungary. ACTA ALIMENTARIA 43. pp.132-206. DOI: 10.1556/AAlim.43.2014.Suppl.19 IF: 0,274

J Soós, Sz Kozits, Z Kovács, E Várvölgyi, D Szöllősi, A Fekete (2013) Application of electronic tongue to beverages. ACTA ALIMENTARIA 42: (Supplement 1) pp. 90-98. DOI: 10.1556/AAlim.42.2013.Suppl.11. ISSN: 0139-3006 (Print), ISSN: 1588-2535 IF:0,427

Articles without impact factor

Ildikó, Magyar and **János, Soós** (2016) Botrytized wines – current perspectives INTERNATIONAL JOURNAL OF WINE RESEARCH 8 pp. 29-39., 11 p.

Conference proceedings in English

János Soós, Evelin Várvölgyi, György Bázár, József Felföldi, Ildikó Magyar, Zoltán Kovács (2015) Electronic tongue in botrytized wine authentication In: 16th International Symposium on Olfaction and Electronic Nose. Dijon, France, 2015.06.28-2015.07.01.pp. 75-76

János Soós, Evelin Várvölgyi, Lajos Dénes Dénes, Sandrine Isz, Zoltán Kovács, József Felföldi (2014) Comparison of two electronic tongue's sensor arrays during wine measurement. AgEng2014 International Conference of Agricultural Engineering, Zurich, Switzerland, 06-10.07.2014. Ref.: P0197, ISBN: 978-0-9930236-0-6

Zaukuu Z. John Lewis; **János Soós**; Ildikó Magyar; Zoltán Kovács. Authentication of Tokaj wines with electronic tongue. 5th ISEKI International Conference, Stuttgart, Germany. 1-6 July, 2018. ISBN 978-3-900932-57-2

Conference abstract in English

John-Lewis, Z. Zaukuu ; **János, Soós** ; Ildikó, Magyar ; Zoltan, Kovacs (2018) Recognition of adulterated aszú wine by the electronic tongue In: Viktória, Zsom-Muha (editor) 2nd International Conference on Biosystems and Food Engineering in memory of Professor András Fekete Budapest, Hungary: Szent István University, Faculty of Food Science