Food Chemistry 150 (2014) 287–295

Contents lists available at ScienceDirect

Food Chemistry

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

Analytical Methods

Wine authenticity verification as a forensic problem: An application of likelihood ratio test to label verification

Agnieszka Martyna [a], Grzegorz Zadora [b,c,*], Ivana Stanimirova [c], Daniel Ramos [d]

[a] Faculty of Chemistry, Jagiellonian University, Ingardena 3, 30-060 Krakow, Poland
[b] Institute of Forensic Research, Westerplatte 9, 31-033 Krakow, Poland
[c] Department of Analytical Chemistry, Chemometric Research Group, Institute of Chemistry, The University of Silesia, Szkolna 9, 40-006 Katowice, Poland
[d] Institute of Forensic Science and Security and ATVS - Biometric Recognition Group (ATVS), Universidad Autonoma de Madrid, C/Francisco Tomas y Valiente 11, 28049 Madrid, Spain

ARTICLE INFO

Article history:
Received 18 February 2013
Received in revised form 21 October 2013
Accepted 24 October 2013
Available online 1 November 2013

Keywords:
Evaluation of forensic evidence
Food products authenticity
Likelihood ratio
Empirical cross entropy
Classification problem

ABSTRACT

The aim of the study was to investigate the applicability of the likelihood ratio (LR) approach for verifying the authenticity of 178 samples of 3 Italian wine brands: Barolo, Barbera, and Grignolino described by 27 parameters describing their chemical compositions. Since the problem of products authenticity may be of forensic interest, the likelihood ratio approach, expressing the role of the forensic expert, was proposed for determining the true origin of wines. It allows us to analyse the evidence in the context of two hypotheses, that the object belongs to one or another wine brand. Various LR models were the subject of the research and their accuracy was evaluated by the Empirical cross entropy (ECE) approach. The rates of correct classifications for the proposed models were higher than 90% and their performance evaluated by ECE was satisfactory.

1. Introduction

Verifying the authenticity of food products is one of the most important issues in food quality control aiming to guarantee the safety and to protect the rights of consumers and producers. A chemical approach to inferring the properties of food products is based on analysis of chemical composition of a particular food product as a unique combination of constituents. Then, either a classification or a discriminant chemometric method can be used to predict the assignment of an unknown food sample described by its chemical features to a group of similar samples. The classification/discriminant rules are first created for samples grouped according to their geographical origins, years of production, producers or brands, *etc.* (Charlton et al., 2010; Stanimirova et al., 2010) and then these rules are used for prediction purposes. Even though such an approach seems straightforward, it requires a delivery of new food quality specifications for different authentic food commodities and a selection of a classification/discriminant chemometric model with a relatively high efficiency, sensitivity and specificity for the problem studied. Therefore, the development of cost-effective procedures for identification of fraudulent products by checking the compliance with the food quality specifications is highly valued. This was essentially the goal of the EU-funded project TRACE – Tracing food commodities in Europe.

The authenticity of food products may be an issue of forensic interest, especially when it involves economic consequences or causes negative health effects. Then, representatives of the administration of justice are interested in answering the question of *what is the value of the evidence of the measurements in relation to the propositions that the analysed sample came from either category 1 or 2*? This problem is known in the forensic field as a classification problem.

A situation in the court is that the prosecutor and the defence have opposite hypotheses e.g. θ_1: a wine is not from Grignolino brand and θ_2: a wine is from Grignolino brand. In general, the prosecutor and the defence think in a sense of the following conditional probabilities – $\Pr(\theta_1|E)$ and $\Pr(\theta_2|E)$, where E describes the evidence (e.g. physicochemical data obtained during analysis of a wine sample, quality specifications). The role of the forensic expert is to evaluate an evidence (E) in the context of these hypotheses. It requires estimation of the following conditional probabilities $\Pr(E|\theta_1)$ and $\Pr(E|\theta_2)$.

The evaluation of physicochemical data (quality specifications) from a forensic point of view requires some knowledge about the rarity of the measured physicochemical properties (quality specifications) in a population representative for the analysed casework – called the relevant population (e.g. the population of wines of a

* Corresponding author. Tel.: +48 12 422 87 55; fax: +48 12 422 38 50.
E-mail addresses: rzepecka@chemia.uj.edu.pl (A. Martyna), gzadora@ies.krakow.pl (G. Zadora), ivana.stanimirova-daszykowska@us.edu.pl (I. Stanimirova), daniel.ramos@uam.es (D. Ramos).

0308-8146/$ - see front matter © 2013 Elsevier Ltd. All rights reserved.
http://dx.doi.org/10.1016/j.foodchem.2013.10.111

particular type). For instance, similar values of particular wine characteristics could be observed in different brands of wines. Therefore, information about the rarity of a determined value of wine characteristics has to be taken into account. For example, the value of the evidence in support of the proposition that the wine sample originated from category 1 is greater when the determined value of these characteristics is rare in the relevant population of category 1, than when this value is common in the relevant population of category 2. In the aim to obtain information about the rarity of the physicochemical data suitable databases should be available. Moreover, it should be pointed out that information about the rarity is not included in most of the discriminant methods, e.g. LDA.

Moreover, it is important that the results of the physicochemical analysis (quality specifications data) of products subjected to authenticity verification made by forensic experts should be evaluated by methods which also allow for including information about the possible sources of uncertainty (e.g. the variation of measurements within the analysed objects, the variation of measurements between objects in the relevant population) and existing correlation between variables in the case of multi-dimensional data.

The evidential value of physicochemical data (quality specifications), taking into account all the mentioned requirements stemming from forensic practice, could be assessed by the application of the likelihood ratio approach (LR), a well-documented measure of evidential value in the forensic sciences. An extensive body of literature exists on the applications of LR in the forensic field (Aitken & Taroni, 2004). The likelihood ratio approach is widely used in the interpretation of data collected in the analysis of glass fragments (Zadora, 2009; Zadora & Neocleous, 2009; Zadora & Ramos, 2010) and in genetics for DNA profiling (e.g. Aitken & Taroni, 2004; Evett & Weir, 1998). It allows for analysis of the evidence (E) in the context of two hypotheses, that the object belongs to either 1° category (θ_1) or the 2° one (θ_2). The LR is defined by the following equation:

$$\mathrm{LR} = \frac{\Pr(E|\theta_1)}{\Pr(E|\theta_2)}. \tag{1}$$

In the case of continuous type data, $\Pr(\cdot)$ are substituted by suitable probability density functions $f(\cdot)$. Values of LR above 1 support θ_1, while values of LR below 1 support the θ_2 hypothesis. The values equal to 1 support neither of them. The higher (lower) the value of LR, the stronger the support for the relevant hypothesis is.

The likelihood ratio approach is a part of the Bayes' theorem expressed in Eq. (2):

$$\frac{Pr(\theta_1)}{\Pr(\theta_2)} \cdot \frac{\Pr(E|\theta_1)}{\Pr(E|\theta_2)} = \frac{\Pr(\theta_1)}{\Pr(\theta_2)} \cdot \mathrm{LR} = \frac{\Pr(\theta_1|E)}{\Pr(\theta_2|E)}. \tag{2}$$

$\Pr(\theta_1)$ and $\Pr(\theta_2)$ are called *a priori* probabilities and their quotient is called the prior odds. Their estimation lies within the competence of the fact finder (judge, prosecutor, or police) expressing their opinion about the considered hypotheses before the evidence is analysed, thus without having any further information in this matter. This opinion may be modified by accounting LR values supporting one of the propositions and delivered by an expert after the analysis of evidence. It is the duty of a fact finder, police, or court to determine whether the objects are deemed to belong to one of the considered categories and this decision is taken based, as mentioned previously, on the results expressed in the form of conditional probabilities – $\Pr(\theta_1|E)$ and $\Pr(\theta_2|E)$, namely posterior probabilities, whose quotient is called the posterior odds.

For every evidence evaluation method it is crucial that it delivers strong support for the correct hypothesis (i.e. $\mathrm{LR} \gg 1$ when θ_1 is correct and $\mathrm{LR} \ll 1$ when θ_2 is correct). Additionally, it is desired that if an incorrect hypothesis is supported by LR value (i.e. LR < 1 for true θ_1 and LR > 1 for true θ_2), then the LR value should concentrate close to 1 delivering only weak misleading evidence. Roughly speaking, according to Eq. (2), it seems to be of great importance to obtain LR values that do not provide misleading information for the court or police. This implies the need of evaluating the performance of the applied methodology for data evaluation, which could be made by the application of the empirical cross entropy (ECE) approach (Brümmer & du Preez, 2006; Ramos, Gonzalez-Rodriguez, Aitken, & Zadora, 2013; Ramos & Zadora, 2011; Zadora & Ramos, 2010).

The aim of this study is to investigate the applicability of the likelihood ratio approach for verifying the authenticity of samples for forensic purposes. For illustration purposes, a set of authentic wine samples described by physicochemical features that belong to three production brands (Grignolino, Barolo, and Barbera) was considered. The assessment of the performance of the applied models was conducted by the empirical cross entropy approach (Brümmer & du Preez, 2006; Ramos et al., 2013).

The aim of the paper is to present LR approach, which could be used when the authenticity of food products is an issue of forensic interest and to show the performance of LDA when the method was applied for the same forensic purpose.

2. Methods

2.1. Wines database

The data subjected to the evaluation process were taken from Forina, Armanino, Castino, and Ubiegli (1986). They were obtained from the analysis of 178 wine samples from 3 brands of Italian wines (59 samples of Barolo (further denoted as BAR), 71 samples of Grignolino (GRI), and 48 samples of Barbera (BRB)). Each sample represented a single bottle of wine. Samples were collected and pretreated in a way conditioned on the type of the subsequent analysis. The applied methods of the analysis were mostly specific for wines analysis such as a group of methods known under common name wet chemical analysis. The rest of the methods involved HPLC, GC, and enzymatic analysis. Schlesier et al. (2009) discuss these issues.

For each sample, 27 parameters were determined and listed in Table 1. All of them represent the commonly determined characteristics of wines for commercial and scientific purposes.

2.2. Likelihood ratio

In this research LR values were calculated for each of the 178 analysed objects (wine samples). Therefore, the data matrix consisted of 178 rows (each corresponding to one of the analysed samples) and 27 columns (each describing one of the determined parameters for the wine samples). Therefore, the data for the sample under classification were in the form of a $\bar{\mathbf{y}}$ vector with the length of 27. A so-called *one-level* LR model (firstly introduced in Zadora (2009)) was applied since there were only single measurements made for each parameter within an object, thus the within-object variability was not available (Zadora, 2009). For the purpose of this study a likelihood ratio (LR) was computed for logarithmically transformed data (i.e. for example $\log_{10}$(Alc), where Alc stands for the original data describing the alcohol content in the samples). A kernel density estimation procedure (KDE) using Gaussian kernels was applied for the estimation of between-object distribution as some of the variables could not be described by normal distribution (see Section 3.1).

The jack-knife procedure was applied for the estimation of suitable population parameters, which implies excluding the object