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Determination of the geographical origin of Italian extra virgin olive oil using pyrolysis mass spectrometry and artificial neural networks

G.J. Salter [a,*], M. Lazzari [b], L. Giansante [b], R. Goodacre [a], A. Jones [a], G. Surricchio [b], D.B. Kell [a], G. Bianchi [b]

[a] *Institute of Biological Science, Edward Llwyd Building, University of Wales, Aberystwyth, Ceredigion SY23 3DA, UK*

[b] *Istituto Sperimentale per la Elaiotecnica, Contrada Fonte Umano, 65013 Città S. Angelo, Pescara, Italy*

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Abstract

Olives were collected from five important regions of Italy, from as many cultivars and locales as possible. For each region a number of samples were produced, representative of the area as a whole. Once collected the olives were washed and processed using standard methods within the ISE olive mill to produce DOC extra virgin olive oils of known region, province and variety (cultivar). These oils were analysed in triplicate by Curie-point pyrolysis mass spectrometry and the spectra collected. Spectra were normalised and sorted according to region. The data-splitting program, Multiplex (A. Jones, D.B. Kell and J. Rowland, Submitted to Analytica Chimica Acta (1996)) was used to sort the spectra into training and test sets split in the ratio 2:1 for Abruzzo:Sardinia and Apulia:Sardinia predictions and a ratio of 1:1 for Lazio:Sicily. Using artificial neural nets with a single output that represented the network's estimation of the geographical provenance as a numeric code, all unknown samples (as triplicates) from an Abruzzo/Sardinia challenge were successfully identified. Samples form a Lazio/Sicily were successfully predicted/separated when the outputs from the network for each triplicates were averaged, and Apulia/Sardinia were predicted with only a single error for each region. This represents the first report in which the precision and discrimination of pyrolysis mass spectrometry has been shown, when combined with artificial neural networks, to allow the discrimination of olive oils by region. © 1997 Elsevier Science B.V.

Keywords: Artificial neural nets; Regional prediction; Extra virgin olive oil

* Corresponding author

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1. Introduction

The importance of the olive industry to Mediterranean countries is huge. World-wide, 9.4 million tonnes of olive fruit is produced per annum, from 805 million olive trees, occupying some 24 million acres of land. Of this, 98% of these trees are in the Mediterranean area. The world olive oil production annually is worth around $2.5 billion [1]. In total 60 million tonnes of seed oil are consumed worldwide every year, 2 million of which are olive oil [2].

The current market is being extended beyond that of traditional importance, partly because of the supposed health benefits of eating olive oil. There is strong evidence that olive oil consumption may reduce the risk of death due to circulatory system diseases [2,3]. Visioli [4] and Galli [5] suggest that this is due at least partially to the natural antioxidants (including the bitter-tasting glycosidic compound Oleuropein) and micronutrients preventing low density lipoprotein from oxidation and so retarding the formation of atherosclerotic lesions.

Martin-Moreno et al. [6] also note that olive oils contain a 'generous amount of antioxidants' and speculate that 'diets high in monounsaturated fats presumably yield tissue structures that are less susceptible to antioxidative damage than would be the case in high polyunsaturated diets.' They identify an inverse correlation between breast cancer and olive oil intake, as do Trichopoulou et al. [7]. The latter also claim that margarine consumption increases this risk. Trichopoulou et al. [8] suggest that olive oil consumption is one of the factors in the traditional Greek diet that aids the longevity of those elderly people in a study group who follow that diet.

Partly as a consequence of these benefits, olive oil commands a much higher price than most other edible oils with extra virgin olive oil and virgin olive oil being considered the most valuable. Italian olive oil, and in particular Tuscan olive oil, is traditionally the most favoured, and therefore attracts a premium. Next harvest (1996–1997 season) the Italian oil producers will provide the consumers with D.O.C. (Denominazione di Origine Controllata) or C.B.O. (Certified Brands of Origin) virgin olive oils (The so called D.O.C. (Controlled Denomination of Origin) classification of Extra Virgin Olive oil is being introduced in Italy according to law 169/1992). Similar provisions can be found in EU regulations 2081/1992 and 2082/1992, governing the D.O.P. (Protected Denomination of Origin) for agricultural food products). These highly priced oils will be obtained from either a single (or a high percentage content from a single) variety or from several varieties growing in a specified region.

The adulteration of extra virgin olive oil by lower grade olive oil is known to represent a severe commercial problem for the marketing of this product, especially within Europe. The two main forms of adulteration are; (i) the addition other (cheap) seed oils such as corn oil or rapeseed oil to high quality (and cost) virgin olive oils and (ii) the packaging of olive oils from one region as those from another which commands a higher price in the market. Firestone et al. [9] reported on a US survey that 4 out of 5 virgin olive oils were correctly labelled, but only 3 out of 20 olive oils. By 1988 [10] only some 17 out of 31 virgin olive oils were correctly labelled, as were 15 out of 26 olive oils.

An additional difficulty in the correct identification of olive oils is the potential large variability, due to variety, location and environmental differences, in the compositional characteristics of 'pure' virgin olive oils; thus, the availability of reliable methods for authentication of the geographical origin of the oils will be crucial.

Official analysis of virgin olive oil involves a series of several determinations of chemical and physical constants that, will be of little or even no use however in the geographical certification of the sample.

This paper presents an overview on a high-resolution spectroscopic/chemometric method under study in co-operation between the Institute of Biological Science of the University of Wales, Aberystwyth (UWA) and Istituto Sperimentale per l'Elaiotecnica (ISE) devoted to olives and olive oil research (Institute of the Italian Ministry of Agriculture).

The method applied, Curie-point Pyrolysis Mass Spectrometry, consists of oil sample pyrolysis, followed by mass spectrometric analysis of the multicomponent mixture obtained, coupled with advanced multivariate data analysis. In order to authenticate oils from different geographical origins successfully, ANNs (artificial neural nets) are trained with oil samples of known origin. In an early study [11], which was performed double-blind, neural networks were trained with the spectra from 12 virgin olive oils, coded 1 at the output node, and with the spectra from 12 adulterated oils, which were coded 0. This permitted the rapid and precise assessment of the adulteration of extra virgin olive oils with various seed oils, a task which previously was labour intensive and very difficult. It was most significant that the traditional 'unsupervised' multivariate analyses of PCA, CVA and HCA failed to separate the oils according to their virginity or otherwise but rather discriminated them on the basis of their cultivar. Training of artificial neural networks is effected by presenting the machine with as many oils as possible that are representative of whole olive growing areas producing D.O.C. oils (an excellent tutorial review has been writen by Goodacre et al. [12] on the use of artificial neural networks on with pyrolysis mass spectra). D.O.C. virgin olive oils are obtained in the ISE olive mill by using the appropriate crushing and related facilities to ensure that all oils have the provenance claimed.

2. Experimental

2.1. Sample collection and preparation

Olives were collected from all regions of study. As many cultivars and locales as possible were collected from within a region and the samples kept separated from each other (i.e. a bulk sample of each geographical region or cultivar was not produced). Thus, for each region a large number of samples were produced, representative of the area as a whole. Once collected the olives were washed and processed using standard methods within the ISE olive mill to produce D.O.C. extra virgin olive oils of known region, province and variety (cultivar).

2.2. Pyrolysis mass spectrometry

1.5 μl of extra virgin olive oils were applied onto iron-nickel foils and the sample allowed to spread over the whole foil surface to give a thin uniform surface coating. Each sample was analysed in triplicate. The main pyrolysis mass spectrometric system used was the Horizon Instruments PYMS-200X ((UWA) Horizon Instruments Ltd., Ghyll Industrial Estate, Heathfield, E. Sussex); for full operational procedures see [13–16]. The Horizon Instruments RAPyD-400 (ISE) was also used.

The sample tube carrying the foil was not heated prior to pyrolysis (heating was found to give memory affects, unpublished data). Curie-point pyrolysis was at 530°C for 3 s, with a temperature rise time of 0.5 s. The data from PyMS were collected over the m/z range 51–200 and may be displayed as quantitative pyrolysis mass spectra (e.g. as in Fig. 1). The abscissa represents the m/z ratio whilst the ordinate contains information on the ion count for any particular m/z value ranging from 51 to 200. Data were normalised as a percentage of total ion count to remove the most direct influence of sample size per se.

2.3. Data analysis

Normalised spectra were sorted according to region and sample, with triplicates being kept together. The data-splitting program, Multiplex was used to sort the spectra into training and test sets. The details of Multiplex, which has been devised in-house, are reported fully elsewhere [17]. The system is based on Duplex, a method for choosing an optimal split between training and test data sets [18]. Multiplex is an extension to this methodology, and splits the data between an

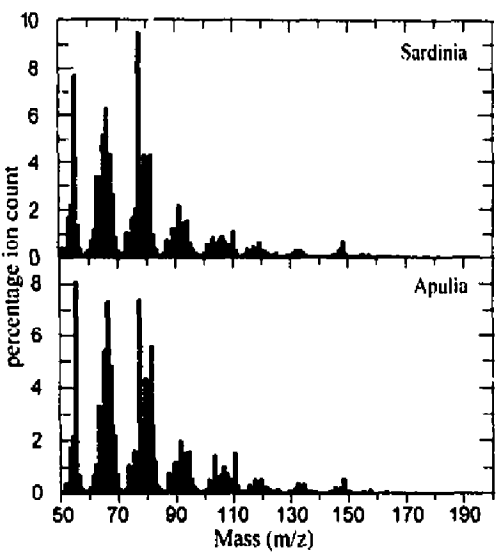

Fig. 1. Typical spectra from extra virgin olive oils.

arbitrary number of sets in a manner similar to duplex. By using this method, we can ensure that models are created on representative data, and tested without fear of extrapolation. A user friendly program has been written that allows splitting on the basis of *X* (inputs, e.g. mass spectra), *Y* (outputs, e.g. region of origin) or both *X* and *Y* data, will read from a number of file formats, and outputs files in a number of formats. In addition, the program will handle replicates and groupings which need to be preserved, and split respectively. The training and test data were split in the ratio 2:1 for Abruzzo:Sardinia and Apulia:Sardinia predictions and a ratio of 1:1 for Lazio:Sicily. So that we were not simply testing the reproducibility of the instrument care was taken to ensure that all three spectra for each triplicate were placed into one group only.

Analysis/prediction was carried out using artificial neural nets (ANNs (NeuFrame version 1,1,0,0 (Neural Computer Sciences, Lulworth Business Centre, Nutwood Way, Totton, Southampton, Hants), which runs under Microsoft Windows NT on an IBM-compatible PC). The architecture is composed of an input layer of 150 nodes, one node for each mass within the spectrum of each oil, a hidden layers of 6 or 8 nodes, and a single output that represents the network's estimation of the geographical provenance as a numeric code; for example Sardinia (code 1) versus Abruzzo (code 0). Headroom was maintained at $\pm$ 10% (for each input) for all networks. A sigmoid (logistic) transfer function has been used as the data relationship is suspected to be non-linear. The supervised learning algorithms was Standard Back Propagation (that updates weights as each pattern is presented) with a learning rate of 0.2 and a momentum rate of 0.8. Patterns were presented randomly with noise of 0.05 added. All data going into and out of the net were scaled automatically (each column individually) by the software.

3. Results and discussion

The separation of many seed oils generally represents a difficult challenge and the methods involved are usually complex [19–23]. The separation of extra virgin olive oils on the basis of the region of origin alone within one country is difficult indeed, and to our knowledge has not been adequately achieved to date by any method. The characteristic spectra produced by PyMS for 'typical' extra virgin olive oils of differing regions are rather similar (Fig. 1), which is not surprising as the oils themselves are too. As can be seen most of the peaks in the spectra are common to all the oils, but in varying concentration (percentage ion count). In fact most of the fatty acids that make up the triglycerides (triglycerides represent over 98% of the whole oil) generally have the same structures, differing mainly in: (a) the degree of unsaturation, (b) the chain length and (c) the relative proportions on the 1-,3- and 2-position of the glycerol backbone. In fact, a single (esterified) fatty acid (oleic acid) may account for 83% of the content of an olive oil [1].

Variations between oils does however exist, and some of these differences will work in our favour and aid the identification of the region of origin, whilst other variations will work against this and make the problem more complex. This

variation arises from a number of sources including the following (amended from Ref. [1]):

1. during the formation of oil in the fruit,
2. between the formation of the oil within the fruit to collection of the fruit,
3. harvest and storage of the fruit.
4. extraction processes,
5. storage of olive oil.

3.1. During the formation of oil in the fruit

Most important amongst these factors are the olive cultivar (the major cultivars for the areas sampled may be seen in Fig. 2), soil, climate and the region where the olives are grown. These all contribute to produce a noticeable difference between oils from the commonly recognised regions. However, within a region highly distinctive locales or provinces may also be found. Additionally climate will also result in differences from year to year within the same region.

3.2. Between the formation of the oil within the fruit to collection of the fruit

Insects, micro-organisms, or other environmental agents, may affect the quality of the oil during the period from its formation within the fruit to the collection of the fruit. Lipolytic enzymes are involved in many undesirable changes. This effect is further complicated by the fact that fruit shaken from the trees deteriorate faster than fruit allowed to fall naturally.

3.3. Harvest and storage of the fruit

The quality of an olive oil is greatly affected by the time of harvest; traditionally this has been at the green-yellow or black-purple stage. Early or late harvests have a negative effect on both the quality and quantity of oil produced. Harvesting itself can take place by (i) hand picking of the fruit from the trees, (ii) after a natural fall of fruit, (iii) by beating the branches or (iv), mechanical shakers. Fruit harvested by hand from the trees undoubtedly yields the best quality olive oil and accounts for up to 20% of the production from Italy.

Often the olives are stored before processing. The most serious damage to the final oil is caused by fermentation of the olives, often caused by poor storage of the fruit within plastic bags.

3.4. Extraction processes

While the extraction of olive oil is essentially a simple process it does consist of several steps (including feeding, washing, crushing, malaxation, separation of the olive oil and centrifuging the oil), many of which could adversely affect the quality of the finished product. Olive oil separation in the past is typically done by either a pressure process, centrifugation or a combination of the two. Although such oils cannot be extra virgin, solvent extraction is a increasingly widely used method for olive oil extraction.

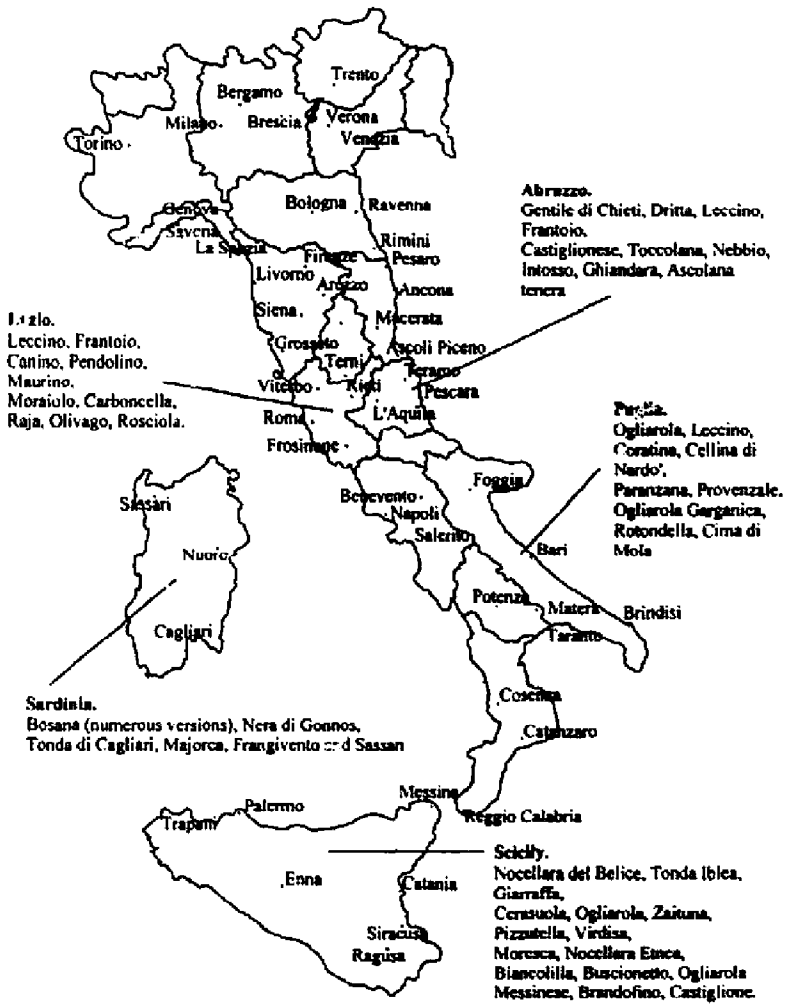

Fig. 2. Map of Italy showing the regions separated and the chief cultivars grown.

3.5. Storage of olive oil

Olive oil deteriorates during storage, the degree being dependent on storage conditions. This is a continuous and irreversible process due to oxidation. Fermentation also occurs in small particles not removed during the filtering of the oil.

Clearly as the olives were processed at the Istituto Sperimentale per l'Elaiotecnica, several of these sources of variation are kept to a minimum; most however will apply.

Discrimination of oils according to region was first attempted by the use of unsupervised multivariate data analysis methods such as principal components

analysis (PCA) and canonical variates analysis (CVA) with very poor results (data not shown). Indicating that these methods could not be used to seperate the olive oils into their different regions.

Initial studies using artificial neural networks (ANNs) to separate olive oils based on the region of origin were more promising. ANNs are a well-known means of uncovering complex, non-linear relationships in multivariate data, whilst still being able to map the linearities. The relevant principle of supervised learning in ANNs is that the ANNs take numerical inputs (the training data) and transform them (usually via summation) into 'desired' (known, predetermined) outputs. The input and output nodes may be connected to the 'external world' and to other nodes within the network. The way in which each node transforms its input depends on the so-called 'connection weights' (or 'connection strength') and 'bias' of the node, which are modifiable. The output of each node to another node or the external world then depends on both its weight strength and bias and on the weighted sum of all its inputs, which are then transformed by a (normally non-linear) weighting function referred to as its activation or squashing function. The great power of neural networks stems from the fact that it is possible to 'train' them (see [24–28]).

Normalised data from Abruzzo and Sardinia were separated into training and test sets using Multiplex. A three-layered ANN (150 input nodes [$\pm$ 10% 'head-room'], 8 hidden and a single output node) was trained using 183 objects (39 Abruzzo (13 individual oils replicated in triplicate) coded 0 and 144 Sardinia (48 individual oils replicated in triplicate) coded 1) as described above. The network training was stopped and the model interrogated periodically using 93 separate objects (21 Abruzzo (7 individual oils replicated in triplicate (squares)) and 72 Sardinia (24 individual oils replicated in triplicate (triangles)) previously unseen by the training net, i.e. the ANN was asked to assign a value between 0 and 1 for each sample as a prediction measure for which region the sample most closely matched.

Although we recognise that other methods of encoding these types of differences are possible [29], points below the 0.25 line are deemed as being Abruzzo, those above the 0.75 line Sardinia. This is a much stricter test than would normally be applied to an output from an ANN; even so all 93 are correctly identified. At the beginning of the learning curve, the ANN learnt to predict over 90% of the samples from both regions very quickly, requiring only a few hundred epochs. The few remaining 'difficult' samples often seemed to be predicted no better or worse as the network proceded to train, only improving after some thousands or tens of thousands of epochs (Fig. 3). The output after 110 000 epochs and a training error of 0.009 RMS may be seen in Fig. 4. Lines representing a network estimate of 0.25 and 0.75 have been drawn in.

Clearly most of the data, both within the test set and the training set will be typical for the region. A small but unspecified percentage of data will be atypical however. The use of Multiplex should ensure that such oils are represented in both the training and test sets, but the numbers available for the net to train on will be very small. If such were the case it would be expected that the 'common' or well represented patterns will be quickly learnt by the net, while the poorly represented oils are either learnt at a slower rate or not at all. An example of where the poorly

represented or 'atypical' samples are not recovered and correctly identified may be seen in (Fig. 5); in this case a single sample in each class (Apulia and Sardinia regions) is predicted wrong. Although this is still a very good result it serves to illustrate the importance of collecting as many samples as possible, while ensuring that they are also representative of the region as a whole.

This point may be seen in Fig. 6. While an ANN did manage to predict the region of origin for Lazio and Sicilian extra virgin olive oils, the results are not conclusive as those shown above. In Fig. 6a it may be seen that the ANN did not achieve a perfect discrimination between the two regions for the individual replicates as previously achieved for different regions (Fig. 4). However, by taking the average of the three replicates for each of the ten samples in the test set, a perfect separation can be achieved (Fig. 6b). In this case, training on only 5 different Lazio extra virgin olive oils (each sample has 3 replicates, giving a total of 15 spectra in the training set) and 6 from Sicily is clearly asking a lot of the methods employed, and is in fact a strong endorsement of the method that it performed as well as it has.

In conclusion, it would seem clear from this study that pyrolysis mass spectrometry combined with the splitting of calibration data using Mutiplex (to ensure that models are produced with representative data), with subsequent analysis by artificial neural nets, is a very powerful method able to discriminate extra virgin olive oils on the basis of the geographical origin.

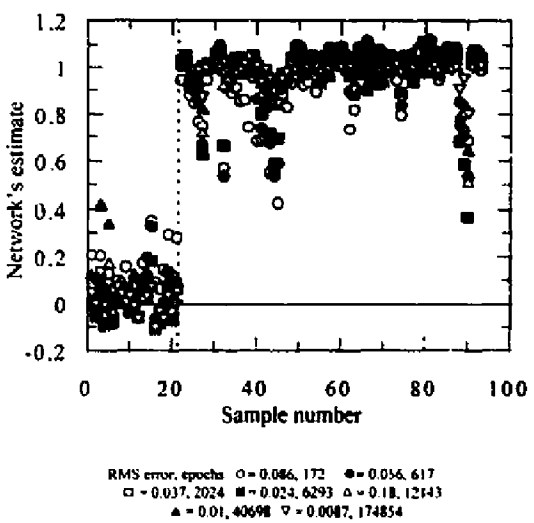

Fig. 3. Training a neural network to discriminate oils from Sardinia and Abruzzo on the basis of their pyrolysis mass spectra. Normalised data were separated into training and test sets using Multiplex. A three-layered ANN (150 input nodes [± 10% 'headroom'], 8 hidden and a single output node) was trained using 183 objects (39 Abruzzo coded 0 and 144 Sardinia coded 1) and interrogated periodically using 93 separate objects (21 Abruzzo and 72 Sardinia) previously unseen by the training net. All samples to the left of the dotted vertical line originate from the Abruzzo region, those to the right Sardinia.

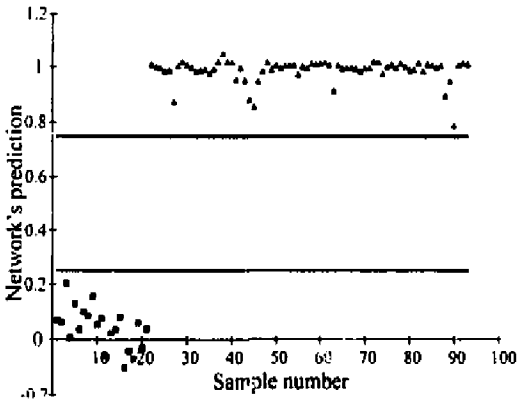

Fig. 4. Testing a neural network to discriminate oils from Sardinia and Abruzzo on the basis of their pyrolysis mass spectra. Normalised data were separated into training and test sets using Multiplex. A three-layered ANN (150 input nodes [± 10% 'headroom'], 8 hidden and a single output node) was trained using 183 objects (39 Abruzzo coded 0 and 144 Sardinia coded 1) and interrogated periodically using 93 separate objects (21 Abruzzo (■) and 72 Sardinia (▲)) previously unseen by the training net. The output after 110 000 epochs and a training error of 0.009 RMS may be seen above. The lines represent a network estimate of 0.25 and 0.75. Points below the 0.25 line are deemed as being Abruzzo, those above the 0.75 line Sardinia. In this strict test all 93 are correctly identified.

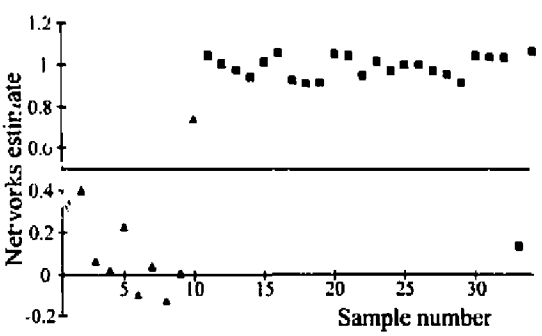

Fig. 5. Normalised data were sorted into training and test data using Multiplex. A three layered ANN (150 input nodes, 8 hidden and a single output node) was trained using 201 objects (57 Apulia coded 0 and 144 Sardinia coded 1) and interrogated periodically using 102 test objects (30 Apulia (▲) and 72 Sardinia (■)) previously unseen by the training net. The output after 5500 epochs and a training error of 0.05 may be seen above. The data are shown as averaged triplicates. The horizontal line corresponds to a net estimate of 0.5; points above the line are deemed as 1 s, and thus a Sardinia sample, and those below as 0 s, and thus Apulia. As may be seen, only one sample within each class is incorrectly identified by the network.

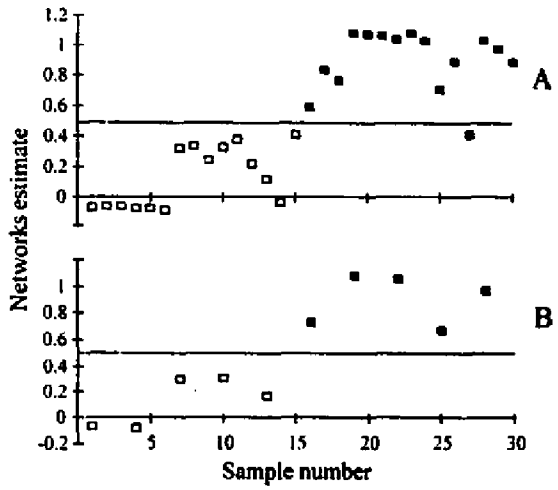

Fig. 6. Normalised data were sorted into training and test data using Multiplex. A three layered ANN (150 input nodes, 6 hidden and a single output node) was trained using 33 objects (15 Lazio coded 0 and 18 Sicily coded 1) and interrogated periodically using 30 test objects (15 Lazio (□) and 15 Sicily (■)) previously unseen by the training net. The output after 8000 epochs and a training error of 0.0196 may be seen above. Graph A shows all samples in triplicate form, B shows the output for each triplicate averaged. The horizontal line corresponds to a net estimate of 0.5.

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