

Use of electronic nose and trained sensory panel in the evaluation of tomato quality

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Abstract: In this paper the performances of an electronic nose based on metalloporphyrin-coated quartz microbalance sensors and of an experienced panel of seven human assessors in the evaluation of gases derived from degradation reactions in tomatoes are presented and discussed. The performances are measured in terms of the capability of both systems to distinguish between samples of different quality coming from conventional and organic production systems. The study deals with the application of pattern recognition techniques based on either multivariate statistical methods (PCA, GPA) or artificial neural networks using a self-organising map (SOM). The response pattern of the sensor array and the sensory data are analysed and compared using these methods. Similarities in the classification of the data by electronic nose and human sensory profiling are found.

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INTRODUCTION

The advent of artificial sensor systems able to mimic chemical senses, such as those known as electronic noses, has opened up a variety of practical applications and new possibilities in many fields where the presence of odours is the phenomenon under control.¹ Many areas in the near future will be touched by these noses, mainly owing to their promising, if still unknown, potential in terms of sensitivities, resolution and stability, and a huge growth of new markets is foreseen. These areas concern indoor and outdoor environmental control, quality assessment of foods and beverages, numerical storage of perfumes, water analysis, skin odour investigation, etc.

Electronic noses are instrumental apparatus based on a chemical sensor array where each sensor is characterised by its own degree of selectivity. This last feature is the key property on which electronic noses found their working principle. Electronic noses are commonly used for classification purposes, since they can distinguish among samples according to various classification criteria. Typical examples are found in the field of food analysis, where sometimes very intuitive classes are adopted according to accepted categories such as freshness or edibility.²

The basic principle is illustrated in Fig 1. The chemical patterns occurring in a certain environment

are 'translated' by the sensors into a response pattern. With respect to the multidimensional chemical pattern the response pattern has a lower number of dimensions, each of which is basically a combination of components in the chemical pattern. The rule of combination, generally non-linear, is given by the selectivity and sensitivity of each individual sensor.

In any application, optimal performance is achieved when the pattern translation process preserves those features allowing discrimination among those classes which are relevant to the particular case. This procedure based on a reduction of dimensions in the patterns has, as a consequence, a reduction of the information content.

The aptitude of electronic noses for the analysis of foods has been tested by measuring sensor sensitivities to a number of odour-active compounds of interest in food quality studies.³ These compounds are representative of numerous classes of chemicals, such as organic acids, alcohols, amines, sulphides and carbonyls. Nevertheless, their performances are still not clearly understood and interpreted and very little is known about the relationship between the response pattern of an electronic nose and human odour perception.

This study was designed to investigate the potential of an electronic nose and an experienced panel of

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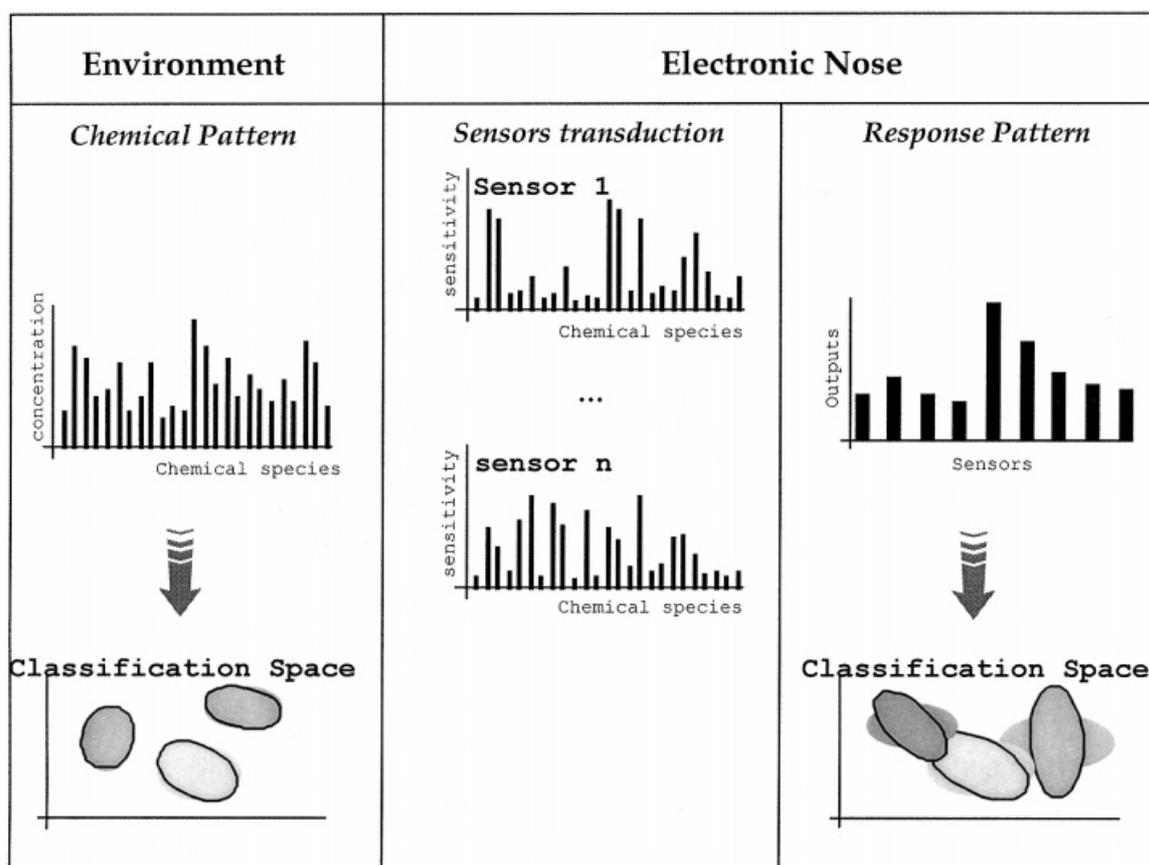


Figure 1. Basic principles of electronic nose. Real patterns occurring in the environment are transduced by the sensor array into a response pattern. In this process there is a drastic scaling of dimensions from the very high number of chemical species present in the environment down to the few units of sensor elements. The electronic nose works properly if the sensor selectivities ensure that in the transduction process those features necessary for correct classification are preserved.

seven human assessors. The case study was the evaluation of undesirable tomato gases derived from sugar metabolism as a consequence of microbial growth. A number of volatile compounds responsible for off-odours, such as diacetyl, acetyl methyl carbinol (AMC), ethanol and acetic acid, can develop as a consequence of mechanical and biological damage due to inadequate harvesting, carrying and storage conditions.⁴

Commonly, organoleptic features of food are checked by human sensory methods. The classical techniques of descriptive analysis, namely flavour profile,⁵ quantitative descriptive analysis⁶ and texture profile,⁷ are useful in the sensory characterisation of a food product. These sensory methods need a group of well-trained assessors using an agreed vocabulary to provide reliable results, requirements that, unfortunately, are time-consuming and, in some cases, could pose serious problems to some industries and laboratories.

In this study the human sensory data and the data from the sensor array were analysed using pattern recognition techniques based on either statistical methods or artificial neural networks. These methods have been chosen in order to determine the similarities between human sensorial and electronic nose ap-

proaches and to investigate the internal classification of both approaches. For this reason, results obtained only by unsupervised methods will be presented in the next sections.

The self-organising map (SOM)

The SOM is one of the most important neural models. It belongs to the category of competitive learning methods and is based on unsupervised learning. This last aspect means that the SOM algorithm does not require any additional information other than the sensor outputs. As an example, an SOM can discover the clustering properties of a set of data without knowledge of their class membership.

The SOM was introduced at the beginning of the 1980s by T Kohonen and, owing to its great flexibility, has been utilised in many different fields, from speech recognition to process control in industrial plants. A comprehensive introduction to the basic principles and many examples of applications can be found in the monograph by Kohonen.⁸ Hereinafter a brief introduction to the SOM is given from the point of view of multisensor applications.

The SOM is a network formed by N neurones most often arranged as a planar grid. Each neurone is identified by a vector \mathbf{r} whose components are the

node co-ordinates in the grid. The neurones are logic elements with two possible states; they have m inputs (vector \mathbf{z}) and one output. An input is a real-valued vector, while the output is either active (value 1) or inactive (value 0).

Each individual neurone is characterised by an m -component codebook vector \mathbf{w}_r which represents the neurone in the input space. In our sensor system this logical structure accepts inputs from the sensor array: according to each input, the codebook vectors of neurones are modified by a learning algorithm, called the 'Kohonen algorithm', which aims at constructing the whole set of codebook vectors $\{\mathbf{w}_r\}$ of the grid as a discrete representation of the distribution of input vectors, ie reference vectors represent areas of the input space that are densely populated.

Once a new \mathbf{z} is provided, the learning algorithm prescribes two stages.

1. *Response*—determination of the index \mathbf{s} from the condition

$$\|\mathbf{z} - \mathbf{w}_s\| \leq \|\mathbf{z} - \mathbf{w}_r\| \quad \text{for all } \mathbf{r} \quad (1)$$

ie the neurone whose codebook vector is closest to the input \mathbf{z} is selected.

2. *Adaptation*—variation of the codebook vectors of all the neurones according to

$$\mathbf{w}_r^{\text{new}} = \mathbf{w}_r^{\text{old}} + \alpha h_{rs}(\mathbf{z} - \mathbf{w}_r^{\text{old}}) \quad \text{for all } \mathbf{r} \quad (2)$$

where h_{rs} (neighbour function) can have for example the form

$$h_{rs} = \exp\left(\frac{-\|\mathbf{r} - \mathbf{s}\|^2}{2\sigma^2}\right) \quad (3)$$

The function h_{rs} defines an area around neurone \mathbf{s} involving those neurones participating in the adaptation stage. The parameter σ is the length scale of the proximity of neurone \mathbf{s} .

From a practical point of view a calibration data set is used to train the SOM, and at each learning step a datum, randomly selected with replacement, is presented to the SOM. This process takes place until the network converges, ie the codebook vectors do not change by more than a negligible quantity. In order to ensure convergence, the parameter α appearing in equation (2) is not constant but is a decreasing function of time operating along the learning process. Various kinds of decreasing laws (linear and hyperbolic are the most common) can be imposed on α .

Once the network reaches convergence, the codebook vectors of the SOM neurones contain the model of the underlying statistical distribution from which the calibration set is drawn. Obviously, the more the sample is representative of the phenomena, the more reliable is the model encoded in the SOM. In order to extract information about the sensory array from the codebook vectors, a proper interpretation of the codebook vectors of neurones is necessary.

The first information that can be extracted from the codebook vectors is the distances between them, namely between the neurones in the pattern space. It is possible to represent this information on the SOM grid by attributing to the segments joining adjacent neurones a grey level (or a colour) whose intensity is proportional to the distance between the connected codebook vectors. The convention adopted is as follows: a dark colour line corresponds to a large distance, while a light colour line refers to a closer distance. This representation gives pictorial information about the clustering of the data. Indeed, light lines are related to clusters, while dark lines are indicators of gaps between the neurones. By fixing a threshold on the distance, it is possible to define clusters on the SOM grid and hence represent the internal classification of the environment by the sensors of the array.⁹ This kind of analysis is similar to that obtainable using simple statistical tools such as principal component analysis (PCA) and cluster analysis. It is worthwhile to discuss the relation existing between the SOM and PCA. PCA is a linear projection of the data set onto a space of reduced dimensions, typically a plane. The projection is unique for the whole domain of the data. In contrast, the SOM is a sort of tessellation of the original space. This can be thought of as a set of local linear models linked together. This representation is of course flexible and can take into account possible non-linearity in the data distribution.

EXPERIMENTAL

Sampling and sample preparation

Fresh tomato samples (*Lycopersicon esculentum*), employed for processing into canned diced or crushed products, were obtained from two firms in the Maremma Toscana (Italy) at the beginning of the processing line. The two produces came from conventional and organic farmed plots; the fruits, growing in separate fields but on similar soil under similar climate conditions, were genetically uniform and were harvested at the same time at approximately the same physiological growth stage.

About 400 fruits (33 kg) from each plot were collected on the processing line after selection and washing and, from a preliminary careful inspection for injury and damage, were classified into four classes of quality, here defined as 'very good', 'good', 'fair' and 'poor'. The class 'very good' consisted of fruits not showing any appearance defects. In the class 'good' were included batches with minor defects, ie bleaching, scars or burns, but no biological or mechanical damage. The class 'fair' comprised fruits with modest biological or mechanical damage but not showing evident rot or splitting. In the 'poor' class were included fruits more extensively damaged by rot or splitting and with major defects in texture.

After classification the fruits were reduced to puree, portioned in 200 g glass boxes and immediately frozen

at -40°C until analyses were performed (within 2 days).

For simplicity, in the following text and figures the four tomato classes will be identified with an Arabic number preceded by the letter B for organic (ie B0, B1, B2, B3) or C for conventional (ie C0, C1, C2, C3) produce.

Sensory analysis. The panel calibration

The sensory quality of tomatoes was assessed by testing the odour sensory perception of critical volatile compounds derived from sugar metabolism by degradation micro-organisms, ie acetic acid, ethyl alcohol, diacetyl and acetyl methyl carbinol (AMC). Other attributes were also selected to provide an overall evaluation of the products.

The sensory panel utilised for this purpose was a selected and trained profiled panel of seven assessors with previous experience in tomato assessment.

Preliminary tests were carried out to improve the ability of the assessors to recognise odour defects and consistently quantify sensory properties. For this purpose, reference material was prepared by adding typical degradation products to optimal taste tomato paste at the threshold concentration (point 1 on the evaluation scale) and at concentrations ranging from 10 to 20 times higher and corresponding to 'evident'

off-odour unacceptable for consumption. The assessors carried out the smelling of the four pure compounds in duplicate at room temperature to assess their sensory properties. They agreed upon reference material for use in both defining the attributes and anchoring the intensity scales (Table 1). These reference samples were given to the panellists during the test sessions.

Measurement procedure

Tomato samples were profiled for nine sensory descriptors referring to colour (intensity, tone, whiteness), flavour (fresh, natural) and biological damage compounds (acetic acid, ethyl alcohol, diacetyl, AMC). Odour defects were labelled from 'nil' to 'strong', with 'zero' representing the best possible product and 'nine' the worst.

Each assessor was required to rate the intensity of each descriptor by placing a slash-mark on unstructured line scales (0–9) using a computerised registration system with a computer system in each booth. In order to eliminate the risk that possible differences in colour/appearance could have an influence on the aroma evaluation, colour and odour descriptors were scored in separate evaluation sessions. All testing was conducted in a climate-controlled taste panel room equipped with individual testing booths and under red

Table 1. Sensory attributes and materials for panel training

Attribute	Scale value	Verbal description	Reference material
Colour intensity	0	Very weak	
	9	Very intense	
Colour tone	0	Yellow/red	
	9	Red	
Whiteness	0	Brown	
	9	Light	
Acetic	0	Not perceived	Optimal taste tomato paste
	1	Threshold value	0.06 g kg^{-1} acetic acid in optimal taste tomato paste
	6	Intermediate	0.6 g kg^{-1} acetic acid in optimal taste tomato paste
	9	Strong acetic odour	
AMC ^a	0	Not perceived	Optimal taste tomato paste
	1	Threshold value	0.04 g kg^{-1} AMC in optimal taste tomato paste
	7	Intermediate	0.2 g kg^{-1} AMC in optimal taste tomato paste
	9	Strong AMC odour	
Diacetyl	0	Not perceived	Optimal taste tomato paste
	1	Threshold value	0.03 mg kg^{-1} diacetyl in optimal taste tomato paste
	6.5	Intermediate	0.3 mg kg^{-1} diacetyl in optimal taste tomato paste
	9	Strong diacetyl odour	
Alcoholic	0	Not perceived	Optimal taste tomato paste
	1	Threshold value	0.8 g kg^{-1} ethyl alcohol in optimal taste tomato paste
	7	Intermediate	16 g kg^{-1} ethyl alcohol in optimal taste tomato paste
	9	Strong alcohol odour	
Natural	0	Artificial off-odour	
	9	Characteristic tomato odour	
Fresh	0	Stale	
	9	Fresh	

^a Acetyl methyl carbinol.

lighting for odour evaluations and cool white fluorescent lighting for colour variables.

The samples were tasted in a randomised complete block design. Two panel replications were carried out on each sample. The temperature of testing was about 22 °C. No information was given to the assessors about the origin of the tomato samples.

The Rome Tor Vergata electronic nose

Since 1995, extensive research has been carried out at the University of Rome Tor Vergata on the exploitation of porphyrins and related compounds for chemical sensors.

Metalloporphyrins are basically assembled by four pyrrole rings linked by methenyl groups to form a macrocycle. This basic structure can be modified by complexing a metal at the centre of the structure and/or linking some peripheral groups around the macrocycle.

Recently, metalloporphyrins have been introduced as coating materials for quartz microbalances to obtain chemical sensors.¹⁰ The main feature of such sensors is the dependence of the sensing properties (in terms of selectivity and sensitivity) on the nature of the central metal and on the peripheral substituents, so that with only a little variation in the synthesis process it is possible to get sensors with different sensitivity and selectivity properties. This flexibility makes this class of compounds of great interest for electronic nose applications.

In this paper an electronic nose formed by eight such sensors is considered. The sensors are coated with the following eight compounds: Ru-*meso*-tetraphenylporphyrin, Rh-*meso*-tetraphenylporphyrin, Mn-*meso*-tetraphenylporphyrin, Co-*meso*-tetraphenylporphyrin, Sn-*meso*-tetraphenylporphyrin, Co-*meso*-tetra-*p*-NO₂-phenylporphyrin, Co-*meso*-tetra-*p*-OCH₃-phenylporphyrin and Mn-*meso*-octamethylcorrole. The selected metals are known to act as catalysts when co-ordinated

to porphyrins. They have been largely used in the past to catalyse, for example, oxidation reactions. Expected selectivities should basically follow the hard-soft acid-base principle, which states that hard acids prefer to bind to hard bases and *vice versa*. Thus Mn(III) species (hard acids) show greater sensitivities to oxygen-based ligands than Co(II) species (intermediate acids), which show relatively higher sensitivities to amines and sulphur-containing compounds. This scheme is very simple, because we ignore the role of the porphyrinato ligand, but previous experiments concerning the sensitivity to alcohols and amines¹⁰⁻¹² have confirmed this preliminary hypothesis.

Sensors have been prepared by coating AT-cut quartzes with a fundamental frequency of 5 MHz. Coatings have been obtained by depositing a specified amount of diluted CHCl₃ solution of the compounds, previously listed, on the surface of the quartz. After the evaporation of the solvent, 50 µg of coating was deposited on the quartz.

Sensors have been housed in a test chamber with a volume of 250 ml. Each sensor is part of an oscillator circuit. In order to maximise the electrodynamic range of the quartz oscillations, the Colpitts circuit has been adopted. Frequency measurement has been accomplished by utilising the frequency counter of a Tektronix 2252 digital oscilloscope, the measurements being supervised by a PC that also collected the data.

The electronic nose has been operated in a real environment without any particular conditioning of the ambient conditions. All measurements reported herein have been performed at room temperature and a relative humidity of 40% under atmospheric pressure.

Electronic nose measurements

Fig 2 shows the measurement set-up. Samples were closed in bottles from where headspaces were continuously transferred into the measurement chamber

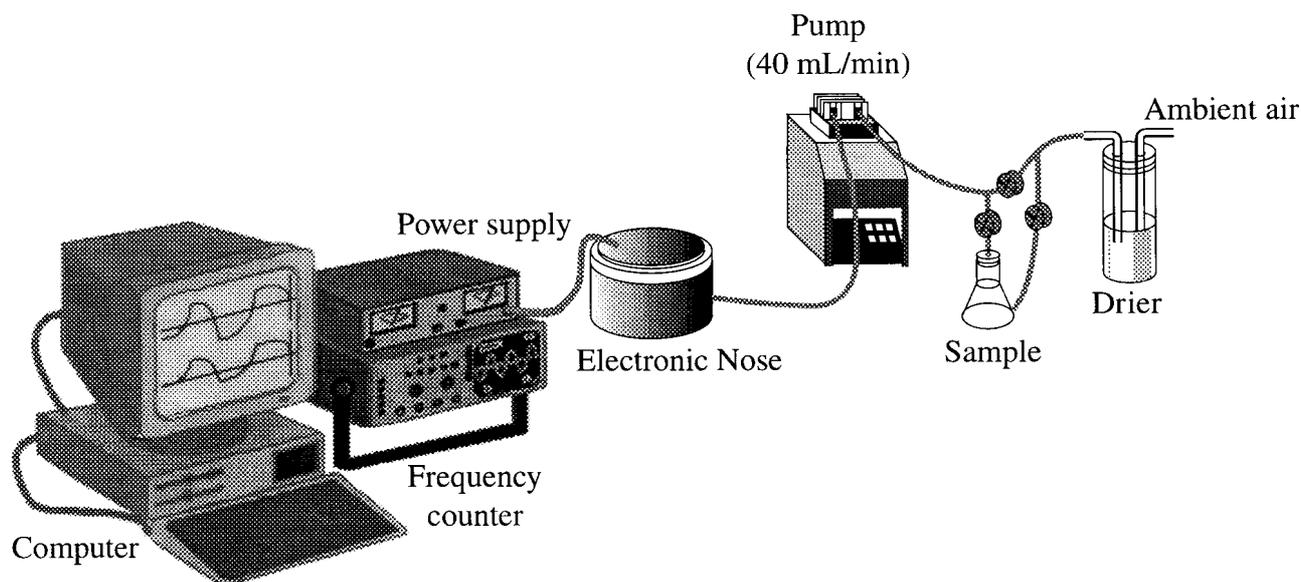


Figure 2. Measurement set-up of electronic nose. As carrier, ambient air is used; the air is dried before coming into contact with the sample.

Table 2. Effect of growing system (S), class of quality (C) and assessors (A) on sensory attributes

	S df=1	C df=4	A df=6	S × C df=4	S × A df=6	C × A df=24	S × C × A df=24
Colour intensity	16.3***	1.9	7.2***	2.1	1.4	0.8	0.6
Colour tone	3.5	2.3	3.9***	0.8	0.9	0.8	0.7
Whiteness	3.2	12.1***	12.0***	8.0***	0.9	1.2	1.4
Acetic	44.9***	16.3***	5.1***	12.5***	1.7	0.9	0.9
AMC	2.9	4.4*	2.2*	1.1	4.1***	1.2	1.1
Diacetyl	61.5***	5.8***	4.7***	8.2***	1.7	0.5	1.4
Alcoholic	1.7	36.5***	6.5***	2.2	2.1	1.4	1.1
Natural	0.0	4.2**	10.0***	2.4	1.6	0.8	0.8
Fresh	0.0	4.2**	10.3***	1.9	0.3	0.9	0.8

*** $P < 0.001$, ** $P < 0.01$, * $P < 0.05$.

by a peristaltic pump. The speed of the pump was chosen in order to maintain equilibrium in the head-space of the sample. Dried ambient air was used as carrier, and measurements were performed at room temperature. In order to test the reproducibility of the measurements, each sample was measured from three to five times.

STATISTICAL ANALYSES

An extensive set of statistical tools for data analysis was available.

A three-factorial analysis of variance (ANOVA 3) with interactions was run on the sensory data in order to analyse the effects of growing system, class and assessor for each sensory descriptor using SPSS.¹³

Owing to their multidimensional structure, electronic nose and profiling data required the use of multivariate techniques for their analysis and interpretation.

The first aim was to determine the contribution of each sensory variable to sensory quality. In order to eliminate the three sources of variation described by Arnold and Williams,¹⁴ namely the level of scoring, the idiosyncratic use of descriptors and the range of scoring, the sensory data set was submitted to generalised Procrustes analysis (GPA)^{15,16} to derive a consensus samples and attributes plot. Procrustes-PC¹⁷ was used to perform GPA. Although principal component analysis is the most commonly utilised method in sensory analysis for data reduction and interpretation, it does not take into account the variances of the product mean scores due to individual differences. Even if the training of assessors can substantially reduce the possibility that different assessors measure different concepts, it is almost impossible to prevent it.

Moreover, linear and non-linear chemometrics-based methods, namely principal component analysis (PCA) and the self-organising map (SOM), have been utilised to analyse electronic nose data in order to extract the relevant information. The same procedures were also followed for the analysis of the sensory data set averaged over assessors. It is worth remarking that the SOM is a neural network which, among many

other features, provides a data representation which is a sort of non-linear PCA, making possible the unveiling of structures which are sometimes hidden in PCA representation. These data analysis techniques have been chosen for their unsupervised character; indeed, although a manual classification of the samples was known, an unsupervised classification places major emphasis on the capability of the electronic nose to detect by itself the different classes, while also providing confirmation of the manual classification scheme.

Data have been compared using these techniques and discussed.

RESULTS AND DISCUSSION

Analysing data from the human assessors

From the ANOVAs, almost all the descriptors except 'colour tone' and 'colour intensity' were significant ($P < 0.01$) to describe the differences among the different classes of quality. Besides, 'acetic' and 'diacetyl' odours also varied between the organic and conventional tomato produces, showing an evident interaction effect. The results are listed in Table 2. Differences among the assessors can be attributed to different average scoring positions on the line scale. Minor agreement among assessors was observed for AMC evaluation ($P < 0.001$) as seen by the growing system × assessors interaction.

The multidimensional structure of the sensory data was reduced by performing generalized Procrustes analysis, which provided a two-dimensional solution explaining 69% variance of the group average configuration (41.4% and 27.6% respectively). Fig 3 shows the sample consensus space and the positions of correlations of original attributes. Ellipses represent the variation around each product point in the space. From the figure the relation between the attributes and the properties of tomato samples can be inferred. The attributes 'whiteness', 'fresh' and 'natural' show a tight positive correlation with the first dimension, whereas the colour attributes 'tone' and 'intensity' and all the volatile compounds which describe the product degradation are negatively correlated with the first dimension.

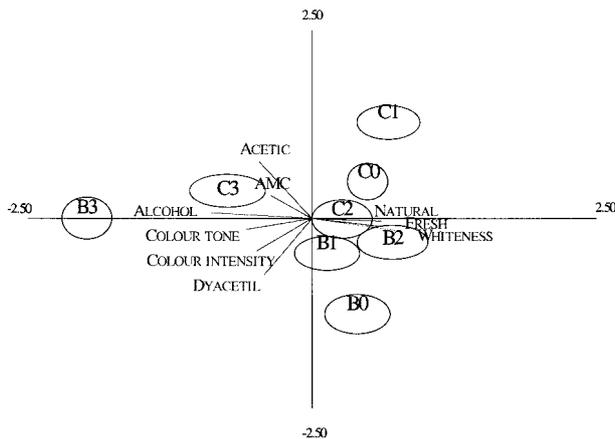


Figure 3. Group average configuration and attribute correlation derived from generalised Procrustes analysis. Experimental data are labelled with the name of their class, as defined in the text.

When dimensions have to be interpreted, the first dimension (x axis in the figure) may be approximately interpreted as a fresh/spoilt axis and describes most of the differences within the sensory data set. Sample positions spread from left to right when the quality improves. It is important to notice the extreme position of class B3 on the negative side in the picture, which includes batches with many instances of mechanical and biological damage. Nevertheless, no consistent pattern emerged for the other classes of quality. The second dimension, explaining a consistent proportion of variance (27.6%), had two odour descriptors more related to it, namely 'diacetyl' (with negative correlation) and 'acetic' (with positive correlation). This second dimension seems to have a role to distinguish between the two types of farming, as also shown by ANOVA. Acetic acid develops as a consequence of the lactic acid bacteria which convert sugar to D- or L-lactic acid and to ethanol or acetic acid. The higher acetic perception in conventional with respect to organic samples could therefore be explained by a higher incidence of damaged batches as a consequence of rougher carrying conditions.

Comparison of electronic nose and sensory data

Electronic nose data and data from assessors have been roughly analysed and compared using principal component analysis (PCA), while a more refined analysis was performed with the self-organising map (SOM) according to a methodology outlined by Di Natale *et al.*¹⁸ Replicate evaluations were treated as different samples in order to have a measure of repeatability.

Fig 4 shows the PCA score plots for both experimental approaches. As can be seen, the score plots exhibit a certain similarity, with class B3 clearly distinct on the right side of the first dimension. The other classes tend to overlap, although the electronic nose data show a certain tendency to be better grouped in the predefined classes.

A more accurate representation of the data can be achieved using the SOM. 10×10 neurone SOMs have been trained using the electronic nose data and the panel data; 21 patterns and 16 patterns were obtained from the electronic nose and panel data respectively. In a recent monograph dedicated to the SOM,⁸ practical rules for the construction of good maps are discussed, but no practical advice is given on the relationship between the number of training patterns and the dimensions of the SOM. In the same reference are shown different samples of maps in which the number of neurones is about six times the number of patterns (Ref 8, p 116). Concerning the application of the SOM to the data analysis of chemical spectra, a ratio of number of neurones to number of patterns of four has been utilised by Göppert *et al.*¹⁹

Fig 5 shows the data of both analyses as they are projected onto SOM grids. It is necessary to remember that the SOM provides a representation of the data which is in some sense a sort of non-linear principal component analysis. It has to be noted that the SOM grid is a discrete space and that the distances between neurones in the original sensor space are not the same, but two neurones which are adjacent on the grid are also adjacent in the original space (topology preservation property). From the figure it is possible to see that

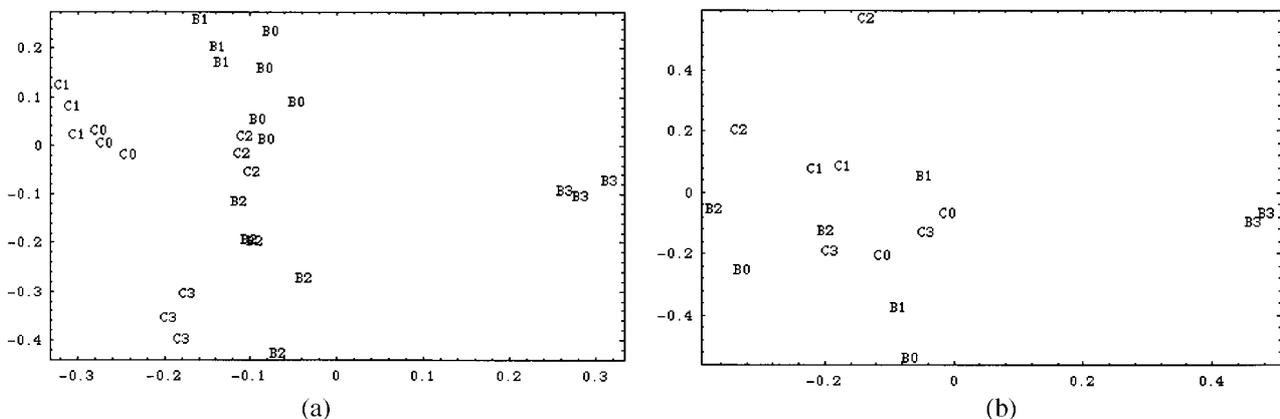


Figure 4. PCA score plots of (a) electronic nose and (b) sensory analysis results. Both analyses show the same conclusions concerning the large difference between class B3 and the rest of the samples. Furthermore, the electronic nose seems to have a better resolution for putting in evidence the existence of similarities between the other classes (eg C0–C1) that does not appear in (b).

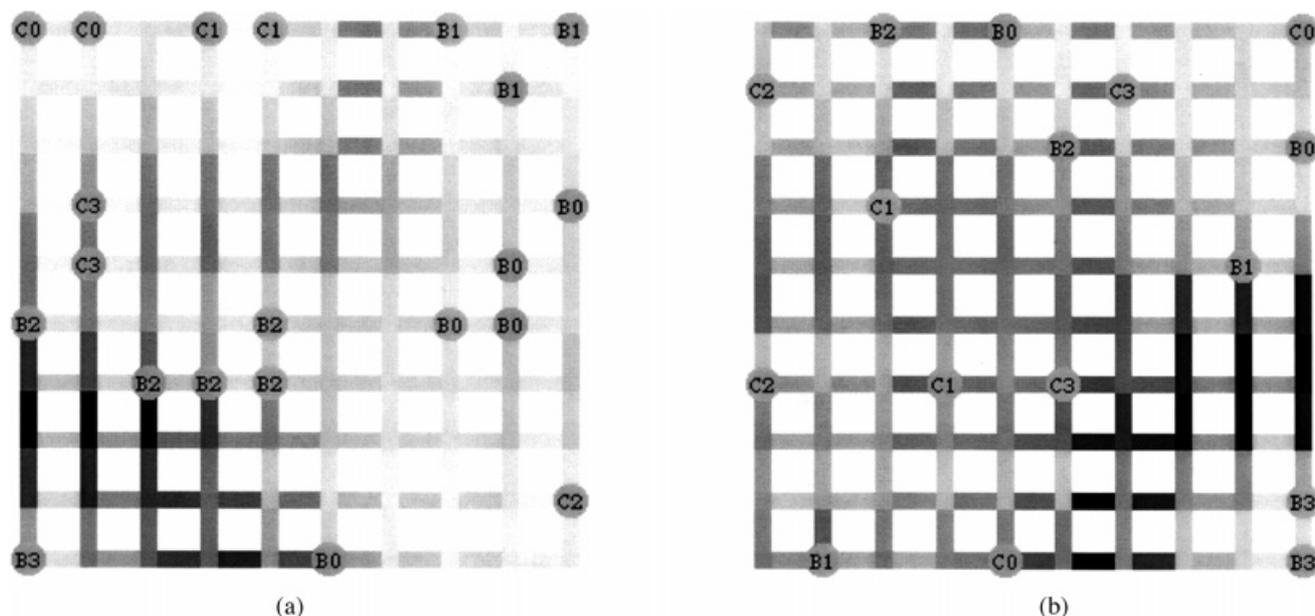


Figure 5. The SOM analysis results are basically similar to the PCA results, with the important improvement that in this case the capability of the electronic nose to correctly classify the tomato classes is evident (a). The same classification is not achieved by the sensory data (b).

electronic nose analysis provides a better-defined class separation and less spread between data in the same class than the sensory descriptive panel.

CONCLUSIONS

It is concluded that both the sensory and instrumental measures were qualitatively valid for the detection of odour defects, but the panel's ability to discriminate between classes close in quality was, in this study, worse than the electronic nose measures; although the variation in sensory data here was higher than usual, their normalisation improves the situation. That is to say, it is necessary that the concentration of an odour change by a factor or two before humans can detect a change in odour.

This work is only a first inspection of the potentiality of the use of an electronic nose for screening and detection of defective tomatoes, which is part of a programme aimed at studying the relationship between the electronic nose and human perception, and does not pretend to exhaust the topic. More work is needed to ensure that the changes noted here are representative.

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