

Evaluation of an Electronic Nose to Assess Fruit Ripeness

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Abstract—The main goal of our study was to see whether an artificial olfactory system can be used as a nondestructive instrument to measure fruit maturity. In order to make an objective comparison, samples measured with our electronic nose prototype were later characterized using fruit quality techniques. The cultivars chosen for the study were peaches, nectarines, apples, and pears. With peaches and nectarines, a PCA analysis on the electronic nose measurements helped to guess optimal harvest dates that were in good agreement with the ones obtained with fruit quality techniques. A good correlation between sensor signals and some fruit quality indicators was also found. With pears, the study addressed the possibility of classifying samples regarding their ripeness state after different cold storage and shelf-life periods. A PCA analysis showed good separation between samples measured after a shelf-life period of seven days and samples with four or less days. Finally, the electronic nose monitored the shelf-life ripening of apples. A good correlation between electronic nose signals and firmness, starch index, and acidity parameters was found. These results prove that electronic noses have the potential of becoming a reliable instrument to assess fruit ripeness.

Index Terms—Electronic nose, fruit ripeness, neural networks, pattern recognition.

I. INTRODUCTION

THE INCREASING competition in domestic and international fruit markets is generating the need for improved ripeness evaluation techniques so that potential losses to the grower and packer, as well as fast spoilage at the consumer end, can be minimized. Although the determination of the optimal timing for harvest and the exact stage of ripeness are among the most important factors in the evaluation of quality in many fruit varieties, the need to find suitable techniques to monitor the ripeness state of a great amount of cultivars still exists [1], [2].

On the other hand, it is well known that a great effort has been carried out to apply electronic noses to the field of food analysis and quality control [3]. Recent papers try to deliver unto

this promise mixing sensing technologies [4] [5] and perfecting signal processing algorithms based on artificial intelligence [6]. Since fruit ripening is associated with an accumulation of aromatic volatiles during ripening for both climacteric and nonclimacteric fruit [7]–[9], electronic noses seem to hold a great potential in the fruit industry. Peaches, for example, seem to be the cultivar with more aromatic volatiles [10].

Some work has already been done on the subject. On the commercial side, hand-held and laboratory instruments have been designed to monitor melon [11] and tomato ripeness [12], and to sort blueberries according to their quality [13]. On the research side, Benady can be regarded as one of the pioneers who used aroma production and semiconductor gas sensors to classify fruit regarding their ripeness state [14]. Since then, many researchers have started to devise systems for fruit monitoring [15]–[20], but rigorous, objective, and well-planned studies for different fruit varieties are only starting to be published now. For example, correlations results between well-established techniques and climacteric fruit, such apples, pears, peaches, and nectarines, have not been published until recently [21], [22].

Our research work in this area included two tasks: to design an electronic nose to measure fruit ripeness and to perform a complete and objective evaluation of the system, comparing the results on different cultivars with those obtained with novel and well-established fruit quality techniques. In order to prove the feasibility of using an electronic nose as a fruit ripeness measuring instrument, the comparison with these techniques is mandatory since, nowadays, although far from perfect, these indicators are the only means to describe fruit ripeness in an objective manner. Therefore, in our studies, we routinely measured fruit sample firmness (which determines how hard the fruit is), colorimetry (to determine the maturity stage through skin color), soluble solids and acidity (through the fruit juice), starch index (a basic measurement for apples), ethylene production (since ethylene is clearly related to the ripening process), and other compounds, such as ethanol, hexyl acetate, etc. These methods are described in greater detail in Section II. Moreover, since ripening is a monotonically increasing process, sensor drift has to be monitored to make sure it does not affect the measurement process.

To be complete, the research in this area has to be applied to different types of fruits, a task that we are doing in our laboratories. This paper describes the experiments carried out so far. Different goals were sought for each cultivar although, in all of the studies carried out, electronic nose signals were correlated with both traditional and novel-quality parameters such as physical-chemical indicators, ethylene production, and aroma quan-

Manuscript received August 9, 2002; revised April 29, 2004. The associate editor coordinating the review of this paper and approving it for publication was Prof. Pavel Ripka.

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Digital Object Identifier 10.1109/JSEN.2004.837495

tification. Hence, fruit quality parameters used to assess fruit quality and maturity were predicted using sensor signals from the electronic nose.

In each study, additional goals were established. In the case of peaches and nectarines, an optimal harvest date for each cultivar was identified from the sensor signal patterns and compared with the optimal harvest dates obtained with fruit quality parameters. In the case of pears, a ripeness classification after different storage conditions was performed using electronic nose signals alone.

The paper is divided in seven sections. First, there is a small introduction on fruit ripeness evaluation techniques. Section III describes the electronic nose prototype designed. The following sections deal with the experiments carried out with apples, pears, and peaches and nectarines, respectively. Finally, section seven outlines the conclusions of our research on the application of electronic noses to the determination of fruit ripeness and discusses the prospects of this new technique.

II. INTRODUCTION TO FRUIT RIPENESS EVALUATION TECHNIQUES

Fruit ripeness is a complex concept that is very hard to quantify or even define. In fact, it is more a perception than an objective quantity. Many techniques have been devised to quantify such a perception. Unfortunately, no single technique presented to date correlates directly with the maturity state of fruit. Different quality measurements have to be made in parallel to get an overall idea of the maturity state of fruit. In fact, fruit professionals use the term "quality indicators" to refer to the parameters extracted when any of these methods is used to monitor fruit ripeness.

Classical fruit quality indicators are based on physico-chemical characteristics of the samples analyzed. These techniques are based on appearance (size, shape, color, and defects), tactile characteristics (firmness), internal characteristics (sugar contents, acidity), or vapor production, such as ethylene [23], [24].

- 1) Firmness is one of the easiest, fastest, and cheapest methods to assess ripeness. It is also one of the parameters that correlates best with ripeness. That is why it is a very popular technique among professionals. It is implemented with a Penetrometer that is sunken into at least two sides of the piece of fruit. The firmness of the samples determines the force that has to be applied to completely penetrate the tip of the instrument into the fruit. In our experiment, it was used in all of the fruits studied.
- 2) Colorimetry is a nondestructive technique that can be applied to determine the state of ripeness of some fruit cultivars. To determine color, a tristimulus Chromameter is used. Parameters such as saturation or hue are described using C.I.E.L *a*b* color space coordinates [25]. This technique is fast, but the instrument is quite expensive. Moreover, it does not correlate very well with the maturity of many fruit varieties. In our experiment, it was used in pears, peaches, and nectarines.
- 3) Soluble solids content (SSC) and titratable acidity (TA) are chemical indicators that are used to assess fruit quality

and ripeness. They are measured in juice pressed from the whole fruit. The SSC can be determined with a refractometer and the TA by titrating juice with NaOH to obtain a pH of 8 and calculating the result as malic acid. Both techniques require the destruction of the sample and do not offer on-line monitoring capabilities. In our experiment, acidity was used in all fruits and SSC on pears, peaches, and nectarines.

- 4) The starch index is rated visually using a 1–6 scale (1: full starch; 6: no starch) after staining an equatorial section of the fruit with a 0.5% I₂-KI solution. Although it is an inexpensive method, it is quite slow and its correlation with maturity is far from perfect. In our experiments, it was used with apples.
- 5) Ethylene is considered a key component of ripeness in climacteric fruit. Its presence activates the ripening process and the ripening process produces more ethylene in climacteric fruit (such as pears, apples, and peaches). Ethylene production is measured by taking samples from the effluent air from respiration jars (where the fruit is placed) that are continuously aerated with humidified air. Sorbent tubes with a chromatograph are used to quantify the ethylene production. In our experiments, it was used with all the fruits.

The state of the art in quality monitoring resides in the indicators that measure the aroma profile emitted by fruit. They are very important from a quality point of view because they are directly related to the flavor and taste of the product when consumed. As an example, Table VIII shows the most important compounds on Big Top nectarines and Royal Glory peaches. Moreover, their presence or absence can be a good indicator of the maturity state of fruit. For example, during fruit maturation, the alcohols (1-butanol, 2-methyl-2-butanol, 1-pentanol, 1-hexanol, and 1-octanol) decrease and the esters (butyl acetate and hexyl acetate), reaching the highest amounts in mature pears [26], [27]. For the extraction of aroma components from intact fruits, the dynamic headspace method can be used.

Finally, additional techniques are being investigated, such as ultrasonics, near-infrared spectroscopy, X-rays, thermography, etc., but all of them pose some difficulties that prevent their use in practical situations [28], [29]

III. ELECTRONIC NOSE DESIGN

A. Original Prototype

We designed a first prototype to measure fruit ripeness [15]. In this initial design, a 50-ml chromatographic syringe (Hamilton, Inc., model 85 020) was used to sample the air inside a sampling chamber (where fruit was placed for a long period of time) and inject the volatiles in the sensor chamber. Although this layout worked, it had two main drawbacks. First, the injection was a manual process subject to a weak repetitiveness and low throughput. Second, the volatile concentration was diluted, since the syringe volume was small compared to that inside the sensor chamber.

Since measurements were time-consuming tasks and volatile concentration was a key issue to succeed in the study, an im-

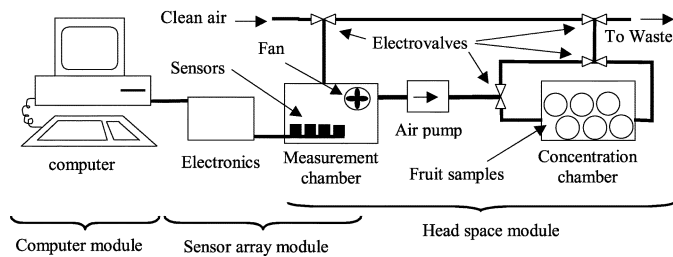


Fig. 1. Schematic diagram of the second prototype.

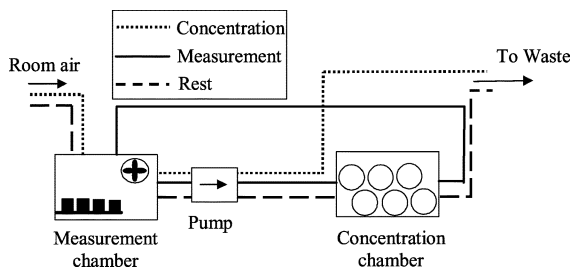


Fig. 2. Air flow paths at different measurement phases.

proved prototype was designed. This is the one described with more detail in the following subsections.

B. Hardware

Fig. 1 shows a schematic diagram of our improved olfactory system, which comprises three basic modules, as follows.

- 1) Headspace module: This module controls the air flow circuit during the measurement process. Inside this module, two chambers, one air pump, tubes, and several electrovalves are included. This module was the main part of the instrument that had to be designed specifically for fruit quality assessment.

The sampling chamber is where fruit is placed. It has a volume of 5 L and its main purpose is to accumulate all the aromatic compounds the fruit releases during the extraction phase. The number of samples depends on their size, but an average of eight fruits were measured simultaneously. The measurement chamber houses the sensor array. Its volume is 1 L.

A pump creates an air flow of 2 l/min. Laboratory room air is used. This decision introduced some interferences, since room air is not controlled and can be slightly contaminated, but the use of laboratory atmosphere reduces costs when operating the system.

During the measurement process, three different phases can be distinguished: concentration, measurement, and rest. The electrovalves, controlled by a computer program, guide the air through different circuits, depending on the measurement phase the system is. No matter the phase, air flow is always kept constant through the measurement chamber. The overall system was in a temperature-controlled laboratory (± 1 °C).

Fig. 2 shows a schematic view of the different air flow paths. When the system is at the extraction phase, air does not cross the sampling chamber, since electrovalves close that path to seal the fruit vessel. The pump gets air

from the laboratory, the electrovalves guide it through the measurement chamber, and, finally, air exits the system. This phase lasted 90 min and was designed to strengthen the aromatic concentration to obtain higher sensor responses. During the measurement phase, the pump pushes the volatiles through a close loop that includes the measurement and sampling chambers. No air enters nor exits the loop. The measurement phase lasts 10 min, time enough for sensors to reach a stable value. Finally, when a measurement is completed, the rest phase is activated. Its main purpose is to clean the circuit and return sensors to their baseline. Room air enters the circuit, crosses the measurement chamber first, the empty sampling chamber afterwards, and pushes the remaining volatiles out of the circuit. Between measurements, a rest time of 15 min was considered appropriate.

- 2) Sensor array module: In this module, we included the gas sensor array, the humidity and temperature sensors, and all the associated electronics necessary to power sensors. The configuration of the sensor array of the electronic nose changed in some experiments. As mentioned before, the sensor array along with humidity and temperature sensors were housed in the measurement chamber. All of the gas sensors that formed the array were semiconductor tin-oxide devices made by Figaro, Inc. and FIS, Ltd. Table I lists all the sensors used and their intended commercial applications. The table also specifies which sensors were actually used in each fruit experiment carried out. Sensitivity to ethylene (which plays a very important role in the ripening process of climacteric fruit) was the initial criteria to recruit sensors for the instrument. Sensor response was evaluated using the relative conductance increment parameter (ΔG_n , described later in detail). Additional tune-up measurements for each application helped to refine the sensor matrix taking into account signal strength (ΔG_n) when measuring each type of fruit. Fig. 3 shows the response of eight Taguchi sensors to a concentration of 10 ppm of ethylene, where it can be seen that when the measurement phase (with ethylene) starts, a change in the resistance of the sensors can be observed. Sensor drift was also monitored and some sensors were discarded for this reason (more on this in later sections). Electronics were necessary to heat sensor elements and to translate resistivity changes into voltage signals the computer could acquire and process. A voltage divider configuration was used as seen in Fig. 4. Power supply and signal conditioning were necessary for temperature and humidity probes also.
- 3) Computer module: The personal computer included in our olfactory system controls the measurement process and, afterward, processes raw data into useful information for the pattern recognition algorithms. With the help of a commercial acquisition board with analog and digital input/output channels, a computer program controls the measuring process. Electrovalves are controlled by binary output signals generated by the program to redirect air flow during the different phases of each measurement. When sensors are exposed to volatiles, during the

TABLE I
ALL THE SENSORS USED BY THE ELECTRONIC NOSE IN FRUIT MEASUREMENTS

SENSORS	APPLICATION	Apples	Pears	Peaches Nectarines
FIS (SB-series)				
SB-AQ1A-00	General purpose. Interior air quality	x		
SB-19-00	Hydrogen	x		
SB-AQ4-00	Cigarette smoke	x		
SB-11A-00	General purpose. Flammable vapours	x		
SB-31-00	General purpose. Organic solvents	x		
SB-30-00	Alcohol. Organic solvents	x		
SB-15-00	Propane/Butane	x		
Taguchi (8-series)				
TGS-800	Air quality. Cigarette smoke, gasoline vapours	x	x	x
TGS-826	Toxic gas detection	x	x	x
TGS-880	Combustible gas detection	x	x	x
TGS-830	Halocarbon gas detection	x	x	x
TGS-825	Hydrogen sulphide	x	x	x
TGS-822	Organic solvents	x	x	x
TGS-2100	Combustible gas detection	x	x	x
TGS-2611	Combustible gas detection	x	x	x
TGS-882	Kitchen control. Alcohol vapours from food	x	x	x
FIS (SP-series)				
SP-MW0	General purpose. Kitchen control	x	x	x
SP-12A-00	Methane	x	x	x
SP-11-00	General purpose. Flammable vapours	x	x	x
SP-MW1-00	Humidity. Kitchen control	x	x	x
SP-AQ1-00	General purpose. Interior air quality	x	x	x
SP-19-00	Hydrogen	x	x	x
SP-AQ3-00	Cigarette smoke	x	x	x

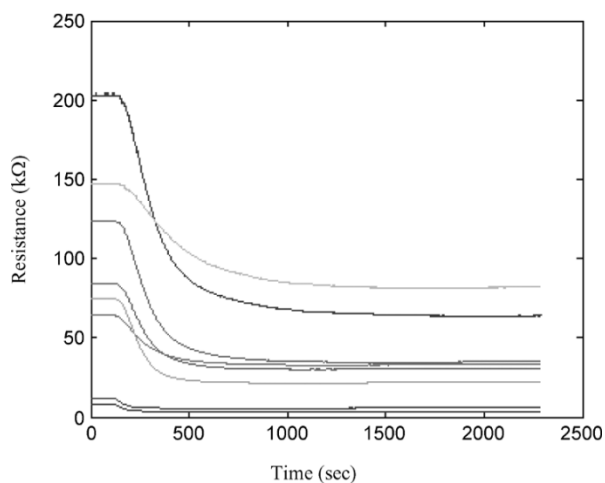


Fig. 3. Sensor responses to 10 ppm of ethylene ($T = 20\text{ }^{\circ}\text{C}$; RH 75%). From top to bottom: TGS-822, TGS-830, TGS-826, TGS-800, TGS-880, TGS-825, TGS-882, and TGS-2611.

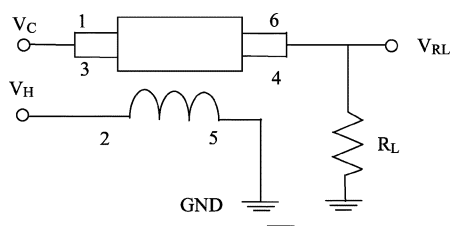


Fig. 4. Sensor electronic connections. Sensitive resistance is located between pins 1 and 3 and 6 and 4. Heater resistance is connected between pins 2 and 5.

measurement phase, the computer records the resistance changes that sensors experience. When a measurement is completed, the acquired data is stored in a hard disk as a

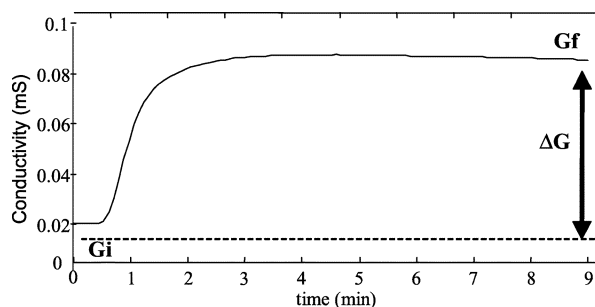


Fig. 5. Response of sensor TGS-800 to fruit aroma ($T = 20\text{ }^{\circ}\text{C}$; RH 75%). The curve represents the conductance transient (mS) of a single sensor under a step change in the concentration of fruit aroma. The graphical meaning of three static parameters (G_i , G_f , and ΔG) is illustrated.

text file for later use. The text file of the recorded measures contains the evolution of the conductivity of each sensor during the measurement (a sample per second for about 10 min). A mathematical package (Matlab, The Mathworks, Inc., Natick, MA) is then used to extract relevant features of each measurement file and to test different pattern recognition methods for each particular goal of the study.

C. Signal Processing

1) *Preprocessing*: In order to apply pattern recognition algorithms, suitable parameters must be extracted from the array of semiconductor gas sensors. Fig. 5 shows a typical response of a sensor (TGS-800) when measuring fruit (eight pears) and the parameters that can be extracted. The curve plotted represents sensor conductivity against time when the volatiles from the fruit reach the measurement chamber due to electrovalve activation. In that transition, the room air flow that

reaches the measurement chamber is substituted by air flow that comes from the sampling chamber, closing a loop circuit between both chambers. It can be seen that, after an initial period of low and stable conductivity (when clean air from the laboratory room is crossing the measurement chamber), conductivity increases sharply and then stabilizes before ending the measurement phase.

From the response of each gas sensor, many parameters can be extracted using MATLAB (see Fig. 5 again). Usual parameters (sometimes called static parameters) include initial conductance (G_i), final conductance (G_f), conductance increment ($\Delta G = G_f - G_i$) and normalized conductance increment ($\Delta G_n = (G_f - G_i)/G_i$). That is why, in each sensor configuration, many parameters were initially used to describe the measurements (more than 50, in some cases). From a scientific point of view, the normalized conductance increment, ΔG_n , should be the best parameter, since it is the one that describes the true relative sensor response toward a given aroma. However, from an experimental point of view, the initial conductance G_i is a very unstable parameter (with low repetitivity) which makes ΔG_n an unreliable parameter. This instability can be derived from the uncontrolled atmosphere in the laboratory combined with the memory effect that sensors suffer if the measurements are not spaced enough time, but spacing measurements too much is not practical since it decreases measurement throughput. The conductance increment ΔG or even the final conductance G_f perform better. The use of PCA [30] and PLS [31] algorithms helped to compress the describing vector for each measurement and obtain results that could be presented in simple two-dimensional plots. For the rest of the algorithms, different combinations were tried, including humidity and sample weight values, that helped to refine the classification results.

The majority of pattern recognition algorithms require some sort of scaling in order to work properly. Even if scaling is not required, it is very useful to apply some sort of normalization due to the different nature of some parameters (humidity, temperature, conductance, etc). In that way, variable relevance is not tied to numerical values that might be very different due to the units used by each variable. Furthermore, linear methods such as PCA and PLS work best with auto scaling. This type of normalization removes the mean value from each variable and then scales dividing them by their variance. In that way, all variables have zero mean and unity variance before feeding the linear algorithms. For this reason, all measurements in our electronic nose were auto scaled when using linear algorithms.

On the other hand, neural networks (like back-propagation, fuzzy art and fuzzy artmap) need input or output values in the [0 1] region. In these algorithms, we scaled measurements dividing each variable by the maximum value found on all measurements. In this way, all the variables were limited to values between 0 and 1.

2) *Unsupervised Pattern Recognition Algorithms*: Unsupervised pattern recognition was used to classify samples without any criteria other than the similarities found by the algorithms. PCA plots helped to see how measurements grouped, whether the electronic nose had enough resolution to discriminate samples, and how variables related to each

other. Moreover, it was also used as a drift detection algorithm when calibration measurements were performed along the experiment.

Fuzzy Art [32] is an unsupervised classification neural network that works remarkably well when there are a few samples to classify. Other advantages include its plasticity to adapt to drift situations and the automatic determination of the number of clusters given a classification criterion (specified with the vigilance parameter). In fact, from a practical point of view, fuzzy art can be considered an automatic and objective clustering algorithm version of a PCA analysis, where clusters have to be manually identified and subjectively drawn. These reasons were considered important enough to include this algorithm in the signal processing toolbox of our electronic nose.

3) *Supervised Algorithms*: To predict analog values (such as fruit quality indicators), a supervised learning algorithm is necessary. In the training phase, the relationships between input and output variables are learned and in the evaluation phase, the rules are applied onto the input values to obtain new outputs (predictions).

Although back-propagation neural networks were initially considered, it seemed inappropriate to use them since they require a large amount of training measurements, something very difficult to achieve when measuring fruit. Partial least squares (PLS), a well-known linear algorithm [31], was considered the best option due to the constraints of the application. During a training phase, the PLS algorithm builds a model that describes the relationship between sensor signals and the fruit-quality parameter to be predicted. In the evaluation phase, the model predicts a fruit-quality indicator using new electronic nose measurements not used for training.

To maximize the use of the measurements and to validate the approach rather than a particular realization of the process, a leave-one-out approach has always been used to predict quality indicators using sensor signals. For a given amount of measurements N , $N - 1$ measurements are used to build a PLS model, while the remaining one is predicted using the model and the corresponding olfactory signals. This process, which is repeated N times (so that each measurement is used once for evaluation and $N - 1$ times for training) optimizes the use of a small set of measurements (the leave-one-out method is sometimes referred to as a cross validation of order one). For each PLS model built, the data used for training are auto scaled; data used for testing are auto scaled using the mean and variance of the training set.

Fuzzy artmap [33], a neural network algorithm, has been included for supervised classification. It has all the advantages fuzzy art has, plus the fact that the classification criteria is specified by the user. In the training phase, fuzzy artmap networks learn from the examples fed to the network. Each example consists of an input vector (with the electronic nose signals) and an output vector that specifies the class to which the measurement should be assigned. In the evaluation phase, sensor signals from a measurement not used for training are fed to the network. Then, the algorithm answers by specifying the class to which the new measurement is closest. In order to validate the approach of using a fuzzy artmap classifier and, to maximize the use of the available measurements during the validation, a leave-one-out procedure was implemented.

IV. APPLE MEASUREMENTS

There were two goals in this study: to see if the electronic nose had enough resolution to follow the ripening process of “pinklady” apples and to look at the correlation between fruit quality indicators and electronic nose signals.

A. Experimental Planning

Two-hundred “pinklady” apples were collected in a single harvest day, the one that was considered optimal by experts on the field. Then, four groups of six apples each were formed randomly selecting samples from the 200 pieces collected (the remaining 176 pieces were used to substitute samples destroyed to obtain quality indicators). At every measurement session, the electronic olfactory system measured separately each group. At the end of each session, nine samples were selected (one from group two, two from group three, and six from group four) for quality analyses. Each piece destroyed was substituted by a new one with similar color, weight, and size, from the 176 remaining samples not chosen initially. Samples from the first group were kept the same until the end of the experiment.

Measurements spanned from day one to day 29 after harvest, keeping the fruit under standard shelf-life conditions (20 °C and 50% to 60% relative humidity). Due to limited resources, measurement sessions were not carried over weekends, and, except for group one, all the groups were measured one time on each session. Group one was measured twice on each session. In all, a total of 88 electronic nose measurements were carried out.

At the end of each session, a calibration measurement was performed. One microliter of ethanol was injected into the sampling chamber and the sensor response was stored. Of course, each calibration measurement was done exactly the same way during the entire experiment, following the same procedure as regular measurements. Time was given to achieve static headspace equilibration. In this way, sensor drift could be monitored during the time period comprised between harvest and the last measurement with pinklady apples.

To assess fruit ripeness, not only the electronic nose, but other more traditional techniques, were applied. In all, from each fruit quality measurement, three quality indicators were obtained. Standard physical-chemical methods applied to determine the ripeness of apples included firmness, acidity, and starch index. These methods implied the destruction of the sample.

B. Classification of Samples by Their Shelf-Life Period

In order to see whether the electronic nose was able to distinguish between different ripeness states, a PCA analysis was applied to the 88 measurements performed with the olfactory system. From each sensor, the ΔG parameter was used, since it is the less likely to suffer from sensor drift.

To check for drift, a projection of the calibration measurements over the fruit measurements PCA was performed. Calibration measurements projected with FIS-SB sensor signals showed a high coincidence with the projections of fruit measurements made the same day. This was a clear evidence that the clustering produced by FIS-SB sensors was artificially created by sensor drift (or other uncontrolled factors) and not by fruit aroma. Similar studies were done with the rest of the sensors,

TABLE II

PREDICTIONS RESULTS ON QUALITY INDICATORS. IDEAL PREDICTIONS WOULD GIVE A ZERO AVERAGE SQUARE ERROR AND INTERCEPT (b), WHILE THE CORRELATION COEFFICIENT AND THE SLOPE (m) WOULD BE 1. THE OPTIMAL NUMBER OF LATENT VARIABLES IS ALSO SPECIFIED

Quality Indicator	Ssq	Corr. coef.	m	b	lv
Firmness	0.1262	0.93	0.91	0	18
Starch Index	0.6	0.68	0.63	0.02	27
pH	0.29	0.84	0.78	0	12

but the projections were scattered in a random manner along the PCA plot. Therefore, we concluded that no significant drift was affecting the experiments when Taguchi and FIS-SP sensor signals were used. Since ambient conditions and the measurements carried out were the same for all sensors, aging was the most probable cause for the drift found in SB sensors. Therefore, FIS-SB sensors were removed from the array and were not used for the rest of the experiments.

C. Correlation Between Electronic Nose Signals and Quality Indicators

As mentioned in Section III-A, some fruit samples from the four groups measured with the electronic nose were used the same day to extract quality parameters (firmness, acidity, and starch index). In order to compare the electronic nose performance with fruit quality techniques, measurements done with the olfactory system were coupled with the values obtained from quality indicators at the same measurement session. In this way, a total of 88 pairs of measurements were coupled.

Table II shows the average square error (Ssq), correlation coefficient, optimal number of latent variables (lv), slope, and intercept for each quality parameter predicted for pink lady apples using the ΔG parameter of all Taguchi and FIS-SP gas sensors. It can be seen that firmness is the best parameter predicted. Fig. 6 shows the prediction ability of the electronic nose for Firmness and pH measurements, where each square represents that predicted against the measured value of each measurement. Ideal predictions would line all points along the diagonal of the plot, where predicted and measured values are the same. It should be kept in mind that measured/predicted parameters were also scaled so that numerical values in the figures do not represent the original values obtained applying the standard physical-chemical methods.

V. PEAR MEASUREMENTS

In this study, the main goal was to classify pears regarding their ripeness state no matter the harvest date or the cold storage period. A secondary study was conducted to see if there was any correlation between electronic nose signals and fruit quality parameters.

A. Experimental Planning

Doyenne du Comice pears were used for the study. At every harvest, a total of 500 samples were collected. Half of them were kept at the fruit research laboratory in Lleida, Spain (where fruit quality measurements were performed), and the remaining ones

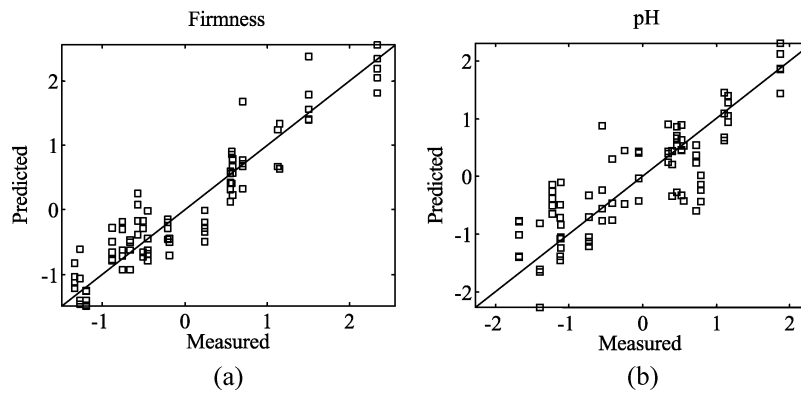


Fig. 6. Firmness and pH predictions using electronic nose measurements against measured values for pinklady apples. Both (firmness in kilograms, pH) have been auto scaled.

were sent to the electronic nose laboratory in Tarragona, Spain. Five different harvest dates were planned.

At both laboratories, samples from each harvest were divided into five groups. The first group was used for shelf-life measurements right after harvest, while the remaining ones were stored inside a cold room. Cold rooms located in both facilities were kept at the same temperature ($1\text{ }^{\circ}\text{C}$) and relative humidity (90%). Each group was cold stored at a different period of time, ranging from one week for group two to four weeks for group five. This distribution was designed to determine the optimal period of time that pears should be stored before reaching consumer markets.

Shelf-life measurements were performed with pears kept at $20\text{ }^{\circ}\text{C}$ during one, four, and seven days after issuing from cold storage. Since there were five harvest dates, five groups for each harvest and three shelf-life measurements for each group, a total of 75 electronic nose and 75 fruit quality measurements were performed in parallel. For correlation purposes, electronic nose measurements were coupled with fruit quality measurements performed on samples from the same harvest and identical storage and shelf-life periods. The same 21 sensors were used (see Table I) and best results were obtained, again, with ΔG .

B. Classification of Samples by Their Ripeness State

In order to see whether the electronic nose was able to distinguish between different ripeness states, a PCA analysis was applied to the 75 measurements performed with the olfactory system. The two first principal components (PCs) captured more than 90% of the variance in the data. Fig. 7 shows the projections of all the measurements onto the two first principal components. Each measurement is represented by a three-digit tag. The first digit identifies the harvest to which samples belonged, the second one specifies the group, and the third digit describes the days of shelf life. In the case of samples from group one (which were not cold stored), the third digit describes the days since harvest.

From the plot, it can be derived that measurements performed on the seventh day of shelf life are fairly well separated from measurements done on day one and day four. Only five measurements performed on day seven are mixed with those performed earlier. These “outliers” belong to the first group of each

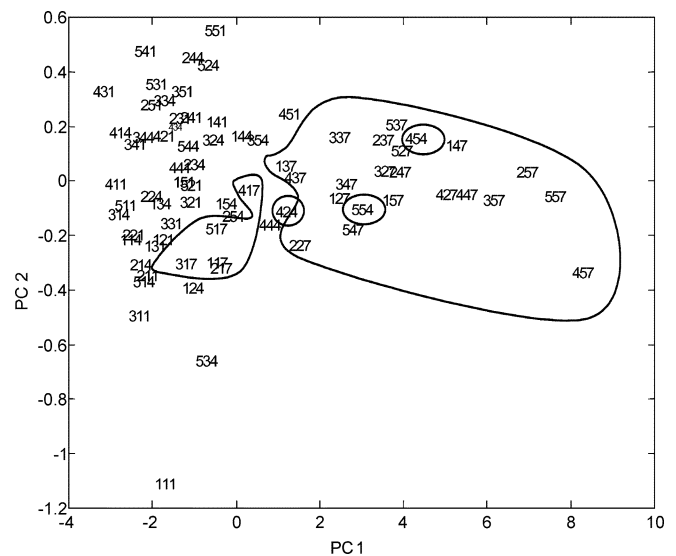


Fig. 7. PCA scores for pear measurements. Solid lines: group measurements of the seventh day of shelf life. Both (firmness in kilograms; butyl acetate in parts per million) have been auto scaled.

harvest, the one that was not cold stored. Literature on the subject [28] states that this pear cultivar needs a chilling period in order to ripen quickly during their shelf lives. Since the electronic nose senses volatiles that are related to ripeness, the “outliers” are, in fact, a clear indication that samples from group one did not ripen at all after seven days of shelf life. Only two samples from day four are located inside the seventh-day cluster and one more is very close. The first two of them belong to the fifth group and fourth and fifth harvest, indicating a more advanced ripeness state. The measurement that is very close to the seventh day cluster can be considered a real outlier since it is from harvest four and group two.

In order to confirm these results, classification algorithms based on fuzzy art neural networks were applied. First, fuzzy art, an unsupervised learning algorithm, was used. The algorithm classified spontaneously the electronic nose measurements in three classes (intended for the shelf-life measurements of day 1, 4, and 7). Most of the measurements performed on day one and four were confined to class 1. On the other hand, most of the measurements performed on day seven were located in class 2 and 3. Table III shows the exceptions to this rule. In this table,

TABLE III
FUZZY ART CLASSIFIED IN CATEGORY 1 MOSTLY ONE AND FOUR SHELF-LIFE MEASUREMENTS, WHILE CATEGORIES 2 AND 3 WHERE MAINLY DEVOTED TO MEASUREMENTS PERFORMED AFTER SEVEN DAYS OF SHELF LIFE. THE TABLE SHOWS THE EXCEPTIONS TO THIS RULE

Class	"Misclassifications"
1 (D1-4)	H1G1D7, H2G1D7, H3G1D7, H4G1D7, H5G1D7
2 (D7)	H4G2D4, H4G5D4, H5G5D4
3 (D7)	None

each measurement is denoted by a string, whose first digit identifies the harvest (H1 to H5), the second one identifies the group (G1-G5), and the final number specifies the shelf-life day of the measurement (D1, D4, and D7). Confirming the PCA behavior, five measurements performed on day seven were located in class one, all of them from group one. Measurements performed on day four were mostly clustered in class 1. Only three "outliers" were located in class 2, the same ones found on the PCA analysis. In fact, two of them belong to the group that was kept longer in cold storage, denoting a tendency to ripen quickly when stored too long. The measurement H4G2D4 is the only one that can be considered a true outlier, just like in the PCA analysis.

Finally, a fuzzy artmap neural network was used to test a supervised classification. The network was trained to classify measurements according to the ripeness state of the samples. Taking into account the conclusions derived from the destructive analyses, two groups were created for the output vector: green or ripen. All measurements done seven days after the harvest were considered ripened, except for group one (samples that did not ripen due to the fact that they were not cold stored). A leave-one-out approach attained a 94.6% success rate (71 measures out of 75). Measurements H4G2D4, H4G4D4, and H5G5D5 were misclassified as ripen and measurement H1G3D7 was misclassified as green. As it can be seen, measurements H4G2D4 and H5G5D5 were the same outliers as in the fuzzy art classification.

C. Correlation of Ripeness Indicators With Electronic Nose Signals

As mentioned in Section III-A, fruit samples measured with the electronic nose were different from samples used to obtain quality parameters. A total of 75 pairs of measurements were coupled, so that each electronic nose measurement was performed on samples from the same harvest, the same identical storage period, and the same shelf-life days than those used to obtain quality parameters.

To see whether the electronic nose was able to predict fruit quality parameters, a leave-one-out approach was performed for each quality indicator. Under this approach, 74 measurements were used to build the PLS model while the remaining one was

TABLE IV
SOME RESULTS ON PREDICTIONS FOR PEAR QUALITY INDICATORS

Parameter	Ssq	Corr. coef.	Lv
Firmness	16.22	0.89	9
Butyl acetate	29.97	0.82	9
Propyl acetate	43.68	0.70	2
Saturation $(a^2+b^2)^{1/2}$	28.51	0.80	7

predicted using the model and the corresponding olfactory signals.

Table IV shows the correlation coefficient, Ssq and optimal number of lv for a few quality parameters predicted for Doyenne du Comice pears, such as firmness and saturation. It can be seen that firmness and color saturation are among the best parameters predicted. Fig. 8 shows a graphical representation of the prediction ability for firmness. Ideal predictions would line all points along the diagonal of the plot, where predicted and measured values are the same. It should be kept in mind that measured/predicted parameters were also scaled, so that numerical values in the figures do not represent the original values obtained applying the standard physical-chemical methods. The best volatile prediction was obtained for butyl acetate and propyl acetate. Fig. 8(b) plots the prediction for butyl acetate and Table IV gives the correlation coefficient and mean square error for two volatile predictions. Details on the chromatographic measurements (procedures and results) can be found elsewhere [26].

VI. PEACH AND NECTARINE MEASUREMENTS

Royal Glory peaches and Big Top nectarines are two cultivars whose optimal harvest date is difficult to predict. That is why the main goal of our measurements was to identify the optimal harvest date using an electronic nose. A secondary goal was to look for correlations between sensor signals and fruit quality indicators.

A. Experimental Planning

Nine different harvest dates were planned. At every harvest date, 60 fruits from each cultivar were collected and divided into four groups of fifteen samples. A first group was assigned to ethylene measurements; a second group was used to analyze aromatic volatile compounds; a third group was used for physical-chemical measurements, and, finally, the fourth group was kept at 20 °C to follow the ripening process with the electronic nose. All of these groups were measured with the olfactory system just after harvest at the electronic nose laboratory in Tarragona. Afterward, the first three groups of each cultivar were sent to the Post Harvest Laboratory, Lleida, where fruit quality measurements were performed. Table V summarizes the experimental plan for each harvest. Table VI summarizes the correlation points obtained for the overall techniques applied. A total of nine correlation points were obtained for Big Top nectarines and nine more for Royal Glory peaches.

Aromatic measurements were performed the third day after harvest and were coupled with the electronic nose measure-

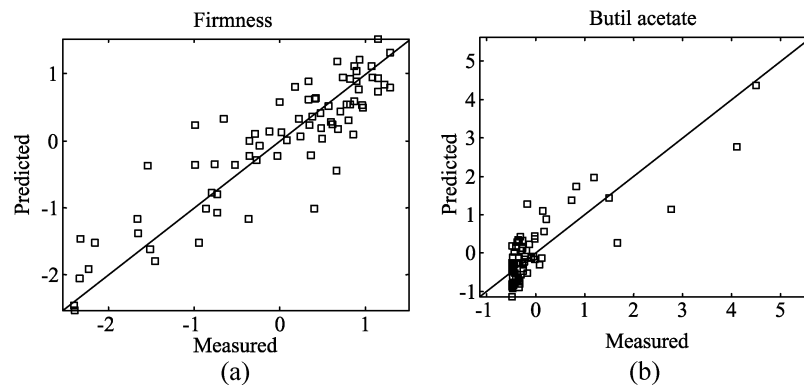


Fig. 8. Best fruit quality parameter predictions for pears. Both (firmness in kilograms; butyl acetate in parts per million) have been auto scaled.

TABLE V
MEASUREMENTS PERFORMED AFTER EACH
HARVEST (VALID FOR BOTH CULTIVARS)

Group	Number of samples	Electronic nose measurements	Ethylene production measurements	Aroma measurements	Physical-chemical measurements
1	15	Harvest day	From day 2 to day 7		
2	15	Harvest day		Day 3	
3	15	Harvest day			Day 4
4	15	From day 1 to day 7			

TABLE VI
CORRELATION CHARACTERISTICS OF EACH MEASUREMENT
TYPE (VALID FOR BOTH CULTIVARS)

Measurement type	Day of measurement	Same day as Electronic nose measurement?	Same samples of Electronic nose measurement?	Measurement points
Ethylene	2-7	Yes	No	24
Aromatic	3	No	Yes	9
Physical-chemical	4	No	Yes	9

ments performed with the same samples the day they were harvested. Physical-chemical techniques were applied to the third group of samples four days after harvest. Again, these destructive measurements were coupled with the electronic nose measurements performed on the same pieces of fruit on the day of harvest.

Since ethylene measurements were performed on group one of each harvest every day during the week after harvest, more correlation points could be used. For correlation purposes, ethylene results were coupled with electronic nose measurements performed the same day with the samples that were kept in the electronic nose laboratory (group four).

In this case, only 12 of the sensors were selected for the experiments. Best results were obtained using both Gi and Gf from each sensor, which means 24 variables were used to describe each experiment.

B. Correlation of Maturity Parameters With Electronic Nose Signals

As mentioned in Section III-A, samples measured with the electronic nose were later used to perform physical-chemical and aroma analyzes. Except for ethylene, nine points could be used for correlation purposes due to the nature of the experiment. To see whether the electronic nose was able to predict fruit quality indicators, a leave-one-out approach was performed for each quality indicator. Under this approach, eight measures

TABLE VII
DESTRUCTIVE PARAMETER PREDICTION RESULTS
ON BIG TOP AND ROYAL GLORY SAMPLES

Parameter	Fruit	Ssq	Corr. coef.	lv
Firmness	BT	1.69	0.94	5
Saturation (most coloured size)	BT	4.3	0.79	4
Hue (less coloured side)	BT	0.3	0.99	7
Firmness	RG	3.5	0.94	6
SSC	RG	4.88	0.92	5
Hue (most coloured side)	RG	1.53	0.98	4

were used to build the PLS model while the remaining one was predicted using the model.

Table VII shows the correlation coefficient, Ssq, and optimal number of lv for some destructive parameters predicted for Big Top samples and Royal Glory peaches. It can be seen that firmness and hue from the less-colored side are the best parameters predicted for nectarines while firmness and hue from the most colored are the best for peaches. Figs. 9(a) and 10(a) show a graphical representation of the prediction ability for firmness for both cultivars.

The best volatile prediction for Big Top nectarines included ethanol and hexil acetate. Fig. 9(b) plots the best prediction (ethylene) and Table VIII gives the correlation coefficient and mean square error for some volatile predictions. Some Royal Glory peach volatiles were also accurately predicted, γ -octalactone being the best one. Fig. 10(b) plots γ -octalactone predictions and Table VIII also includes some Royal Glory results. Detailed description of the corresponding chromatographic measurements and results can be found elsewhere [26].

C. Determination of Optimal Harvest Dates by Unsupervised Learning Algorithms

As mentioned before, the main reason for choosing Royal Glory and Big Top cultivars was based on the difficulty to determine their optimal harvest dates. In order to see if the electronic nose was able to give a good estimate of the optimal harvest date, we tried to classify all the measurements performed with the olfactory system the same day the fruit was harvested.

Fig. 11(a) shows the PCA plot for the two first PCs for Big Top samples. A total of 36 measurements are represented, four measurements for each harvest date (from harvest one to nine). It can be seen that measurements that belong to the harvest one, two, and three groups on three clusters, with low variability

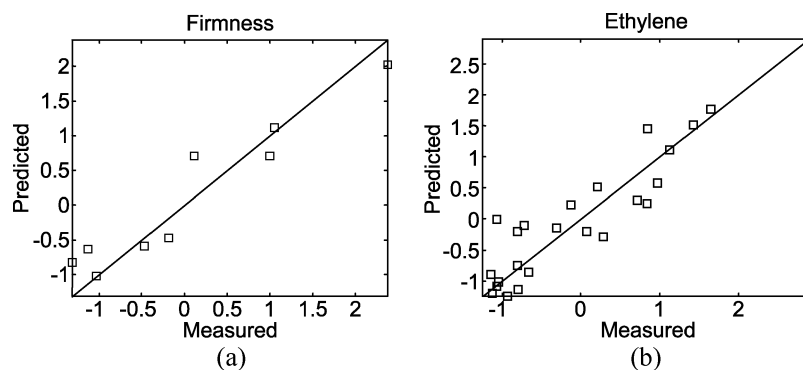


Fig. 9. Two parameter predictions on Big Top nectarines. Both (firmness in kilograms; ethylene in parts per million) have been auto scaled.

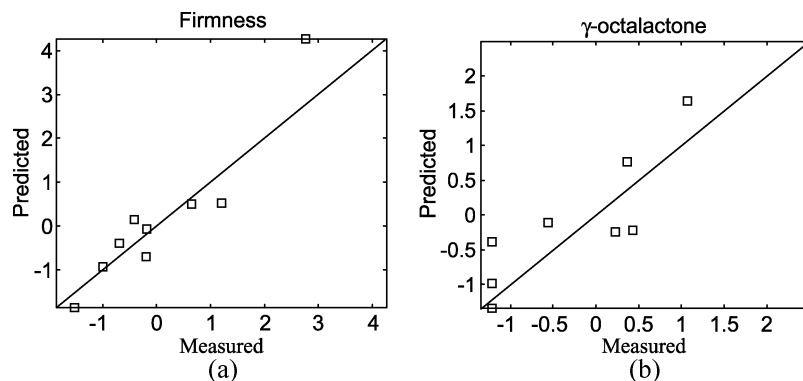


Fig. 10. Two parameter predictions for Royal Glory peaches. Both (firmness in kilograms; γ -octalactone in parts per million) have been auto scaled.

TABLE VIII
AROMATIC PREDICTION RESULTS FOR BIG TOP AND ROYAL GLORY SAMPLES

Aroma	Fruit	Ssq	Corr. coef.	lv
Ethanol	BT	0.42	0.98	8
Hexyl acetate	BT	3.21	0.88	4
(Z)-3-hexenyl acetate	BT	1.15	0.96	6
Hexanal	BT	1.097	0.96	6
1-Propanol	RG	3.84	0.87	6
Ethyl butyrate	RG	4.28	0.85	7
Limonene	RG	8.98	0.72	8
γ -octalactone	RG	2.13	0.91	4

found within each cluster. Measurements done on groups harvested later do not cluster clearly because they show a high variability. The same PCA analysis was performed on Royal Glory samples [Fig. 11(b)] where a similar behavior can be observed, although not as clearly as previously. In this plot, the spreading of measurements occurred after the first harvest.

The same pieces were used to determine the optimal harvest date using physical-chemical and aromatic measurements. Big Top samples started their ripening process between harvest three and harvest four, so the optimal harvest date should lie between both dates. Royal Glory samples started their ripening process after the first harvest, so this was considered the optimal date for Royal Glory peaches.

The PCA patterns turned out to be a key argument to determine at which date the ripening process started using an electronic nose. Since ripening does not occur exactly at the same time for all the samples, a high variability on volatile production should be expected when some fruits started to ripen, whereas no variability should be observed when all samples

were immature. The high variability shown on the PCA plots starts with measurements of the harvest dates found optimal by fruit quality parameters [26]. Therefore, we conclude that the electronic nose detected the optimal harvest dates, especially for Big Top samples.

VII. CONCLUSION

In our work, an application-specific sensor system designed to measure fruit ripeness has been implemented and tested. Studies with apples, pears, peaches, and nectarines, where electronic nose measurements have been correlated with well-established fruit-quality techniques, have shown that some quality parameters can be predicted reasonably well using electronic nose signals without destroying the fruit. Although it may seem surprising to see that physical measurements, such as firmness, can be predicted with sensor responses to organic volatiles generated by fruit, such results are meaningful since the physiological characteristics of fruit are closely related to chemical processes that take place during the ripening process. For example, the electronic nose does not measure firmness directly; it actually measures volatiles that are well correlated with the firmness of the fruit. This is also true for the remaining well-predicted indicators.

Determination of optimal harvest dates with an electronic nose seems to be feasible in some fruit cultivars. To do so, a statistically representative sample of the population of the cultivar has to be chosen, collected, and measured with the electronic nose at the facility where it might have been installed. It is important to stress that great care should be taken on the design

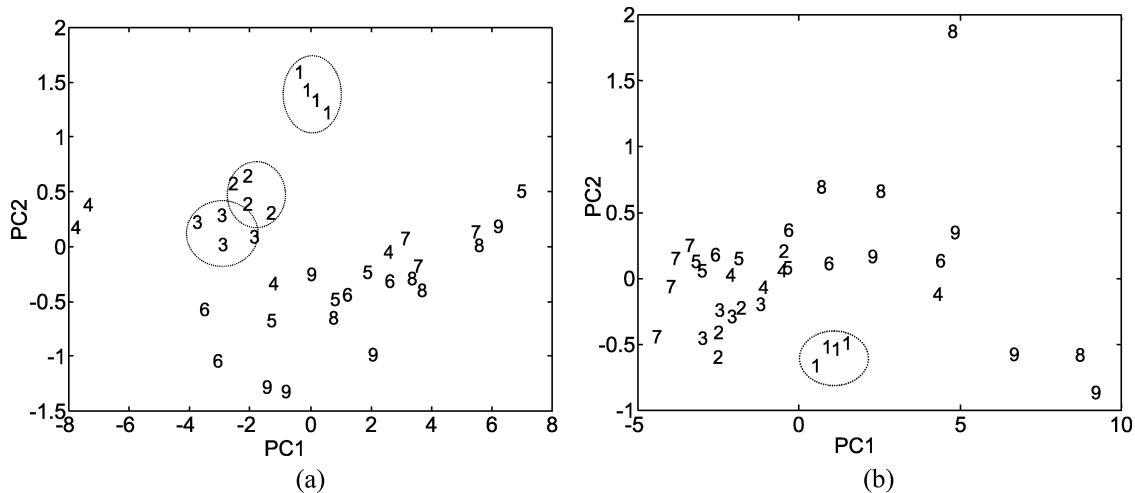


Fig. 11. PCA plot (PC1 versus PC2) for (a) Big Top nectarines and (b) Royal Glory peaches. Harvests are represented by ordinal numbers.

of the sampling procedure (size and methods) to obtain a good prediction on the optimal date of harvest for the entire crop.

Compared to classical and other novel analytical methods, the electronic nose built offers a cheap and nondestructive instrument that (if properly programmed and automated) can be operated by nonspecialists. The number of measurements that can be done in a day compares favorably to other sophisticated methods, such as aromatic profile identification using chromatography (one of the newest approaches), and since the whole process is automatic, the cost of each measurement is very low.

Correlation coefficients quantify the exactitude of the quality indicators predictions. Therefore, in the near future, the electronic nose could be envisaged as a global measurement system calibrated for ripeness determination or a multinstrument system to extract the indicators for which it has been calibrated.

Anyway, further work needs to address important limitations. For example, a straightforward procedure should be devised to detect and correct sensor drift from year to year. Also, the initial calibration of the system for a given cultivar should take only a few measurements and be valid, at least, for some consecutive campaigns. Finally, the measurement cycle should be faster in order to increase throughput. All of these considerations are being studied and might imply the optimization of the sampling process, the use of more advanced processing algorithms, and the incorporation of new sensor technologies into the system.

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