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Olfactory Imaging: An Electronic Nose Using Tiered Artificial Neural Networks And Quartz Piezoelectric Gas Sensors¹.

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Abstract: *Our group is interested in the potential use of gas sensors for detection of key odorants in industry and as alternative sensory mechanisms for guidance control in robots. It has recently been shown that chemically modified, resonating quartz piezoelectric (QPZ) crystals exposed to different odorants, generate characteristic response patterns (termed "kinetic signatures"). To demonstrate their utility in an electronic olfactory system, artificial neural networks (ANNs) were trained using the back propagation method, to associate the kinetic signature responses of 6 differently treated sensors to 18 trialed odorants. Arranging each of the separate networks corresponding to each sensor in a layer-like fashion (hereafter referred to as tiers to avoid confusion with network layers), the weight states of the output processing elements (PEs) of the amalgamated ANN combine to produce weighted, two dimensional 'olfactory response maps' that uniquely identify each of the odorants. Using simple image processing techniques, we discuss how these response maps can give an automated system a degree of feedback as to its physical state, allowing it to detect and potentially rectify problems encountered during normal operation.*

Keywords: artificial neural network, back propagation, gas sensor, quartz piezoelectric crystal, odorants, olfaction.

1. Introduction.

Odorants form a large collection of small organic molecules that are capable of interacting with specific receptor proteins in the mammalian nose [1]. Natural smells (e.g. perfumes and flavours) are extremely complex and can consist of hundreds of different odorant molecules. With the best human noses capable of distinguishing only a handful of pure odorants within any given smell, there exists great potential for electronic noses in industry where sensory analysis of products is currently performed by humans.

Considering that just about every object exudes from its surface airborne molecules that are in most cases indicative of its internal state, then it becomes evident that the potential scope for electronic noses is enormous. In the food and beverage industry for example, an electronic nose could be used to determine the freshness of a product by detecting characteristic odorant

molecules emitted by spoiled produce. Similarly electronic noses can be used to analyse numerous gels, foams and lubricants giving wide application in a number of manufacturing industries. Other possibilities include pollution monitoring, the detection of toxic and odorless gases in the workplace, petroleum products, plastics, narcotics, explosives, biomatter - the diversity of application is enormous.

Despite their promise, electronic noses have not found widespread acceptance for a number of reasons; one of the most pervasive is the volatile nature of the medium in which the odorant sensor must operate. As this can contain many thousands of different gaseous molecules, there exists ample potential for damage of the chemically sensitive sensor by adverse reactions with volatiles in its immediate environment. Consequently the response of the sensor can become highly variable and as such, unpredictable.

Recently a new form of analysis has been introduced that effectively minimises variability in

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the response of QPZ sensors [16]. Complementing this with a novel artificial neural network (ANN) configuration that enables higher order processing using response maps generated from the ANN, a new variant of electronic nose is introduced.

2. Odorant recognition using quartz piezoelectric crystals.

Chemically modified QPZ crystals have been used for many years for the detection of gases and odorants (for current reviews, *see* [11,19]; as biosensors, *see* [18]). At the turn of the century it was known by manufacturers of radio, that the resonant frequency of an oscillating crystal could be decreased by sputtering small quantities of mass onto its surface. It was not however until Sauerbrey [17] derived a mathematical expression; showing that the loss in frequency of a resonating AT-cut crystal operating in the shear mode (the type commonly found in digital circuits), was inversely proportional to the mass loading on its surface, that the potential use of these devices as very sensitive microbalances was realised (for a modern treatment on the crystal dynamics of chemically loaded QPZ sensors, *see* [8]).

Coating the crystal with a chemical substrate that is known to react reversibly with a target gas, the chemically modified QPZ crystal can be used as a gas sensor for that particular gas. The basic operation of the QPZ chemosensor is illustrated in figure 1 using a simple 'lock' and 'key' analogy: A substrate is covalently bonded to the crystal surface whose resultant surface molecular topography presents a series of shaped 'locks', into which only specific molecular orientations of target odorants will 'fit' (I).

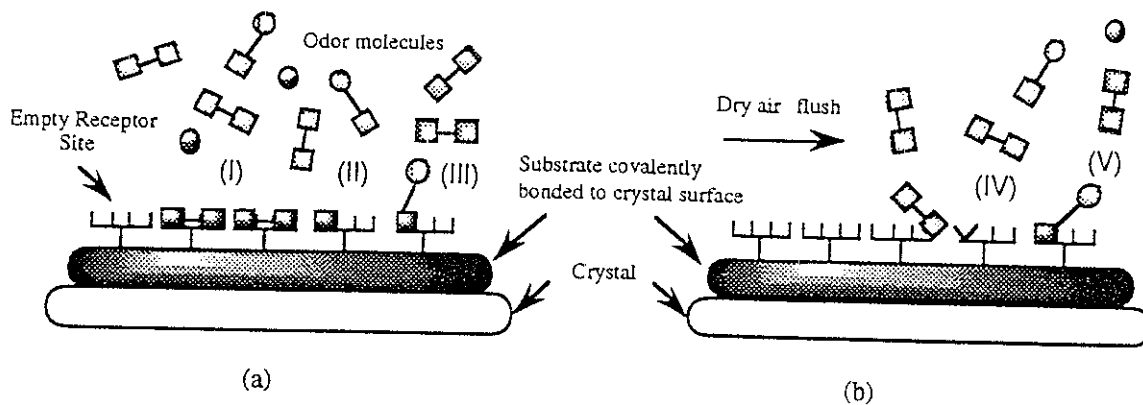


Figure 1. Simplified 'lock' and 'key' analogy of processes occurring during adsorption (a) and desorption (b) of odorant molecules at the sensor surface. During odorant stimulus (a), target odorant molecules ideally attach to their respective receptor molecules (I), although other molecules may also attach (II, III). Following the removal of the odorant stimulus, odorant molecules desorb from the sensor (b), in some cases the original surface may be altered either by molecular change at the surface (IV) or irreversible reactions with odorants (V).

Early users of QPZ sensors used this analogy to target specific gaseous molecules, using the measured loss in resonant frequency of the oscillating QPZ crystal to obtain measurements as to the quantity of target molecules sorbed onto the crystals coating. However because of the huge diversity of chemicals and chemical orientations likely to be found in any given gaseous environment, it is likely that some molecules may 'fit' a part of the lock (II), or have as part of their chemical constituency parts therein that match the lock (III).

To accommodate the lack of specificity encountered in the operation of these devices, modern operators use arrays of differently coated crystals, and codify the odorant-induced peak frequency response of each sensor into a vector that uniquely identifies each odorant. Correct association of sensor responses with odorants is then performed using either statistical techniques [2,3], or more recently ANNs [5,7,12,13].

For sensors to work effectively over extended periods of time however, the selected coatings must react reversibly with the odorants of interest (i.e. after exposure the crystal must return to its baseline frequency as the odorant molecules desorb). A substrate that will react selectively with only one chemical out of all those present in an ambient environment has yet to be satisfactorily demonstrated [11], and inevitable sensor degradation due to modification of the sensors surface (IV) and/or non-reversible reactions with odorant molecules (V) means that the reproducibility of these sensors deteriorates. A number of other factors further detracting from sensor repeatability are: anomalies in sensor coating procedures, moisture adsorption, substrate displacement, and

physical differences in the fabrication of individual crystals [10]

Variability in sensor response is clearly a problem if the peak frequency response of the sensor is to be the only means of analysis, sensors may deteriorate at different rates, and the ability of any pattern recognition scheme to accommodate such change will be limited.

3. Kinetic signatures: A new method of analysis.

Recently a means of representing sensor-odorant interactions that accommodates variability both between identically treated sensors, and sensors of varying efficiency to consecutive exposure of the same odorant has been demonstrated [16]. This form of analysis is made possible by delivering the odorants to the sensor in a flow system that controls odorant concentration, flow rate and exposure time [10]. A sample response obtained from an OV-17 coated sensor to 25 consecutive exposures of ethyl acetate odorant is illustrated in figure 2. On exposure to the odorant, the crystal frequency decreases as the odorant molecules affix to the sensor coating. The amplitude of response and 'shape' of the ensuing response curve, being determined by the particular interfacial kinetics of the odorant coating pair.

Once the odorant stimulus is removed from the sensor, the frequency increases back towards the baseline frequency as odorant molecules desorb from the sensor. Note that the original baseline frequency is not attained for modification of the original surface occurs to various degrees in most reactions observed and only a few odorant-coating combinations react reversibly (in ambient conditions this is practically impossible [11]). The immediate consequence is to effect the responsivity of the sensor to subsequent applications of the odorant.

The current metric used in QPZ sensor

applications is the peak frequency in response of the sensor (Δf_{\max}). Because of the demonstrated variability in responsivity of these devices [10], it is not a measure ideally suited to systems that require a long sensor life, and a more durable means of representing the interaction of sensor and odorant is required. By normalising the response curve $f(t)$ of the sensor to its new baseline frequency f_b following exposure, such that:

$$S(t) = \frac{f(t) - f_b}{\Delta f_{\max}} \quad (1)$$

the kinetic signature, $S(t)$ for each odorant-sensor pair is obtained. These kinetic signatures remain remarkably consistent for all observed reactions where there is pronounced sensor response, despite variation in the responsivity of the sensors to repeated odorant exposure [16]. The corresponding kinetic signatures of the 25 exposures illustrated in figure 2a, are displayed in figure 2b, demonstrating an extremely high degree of repeatability with the exception of the first response (The initial response of some observed reactions was found to be different from subsequent responses. The reason for this is uncertain, but we believe the coating material may react initially with the odorant molecules, in such a way that the original coating is altered and a new, 'conditioned' surface remains. Variability in the initial response may also be due to reactions with surface contaminants (principally water molecules) that have affixed themselves to the surface of the sensor during transit to the gas flow chamber).

To test the suitability of this new form of analysis for an electronic nose, experiments were performed on 18 different odorants (listed in table I), using the kinetic signature responses of three consecutive exposures obtained from 6 differently coated sensors (the chemical coatings used were pyridoxine hydrochloride, antarox CO-880, pyridoxine hydrochloride + antarox CO-880,

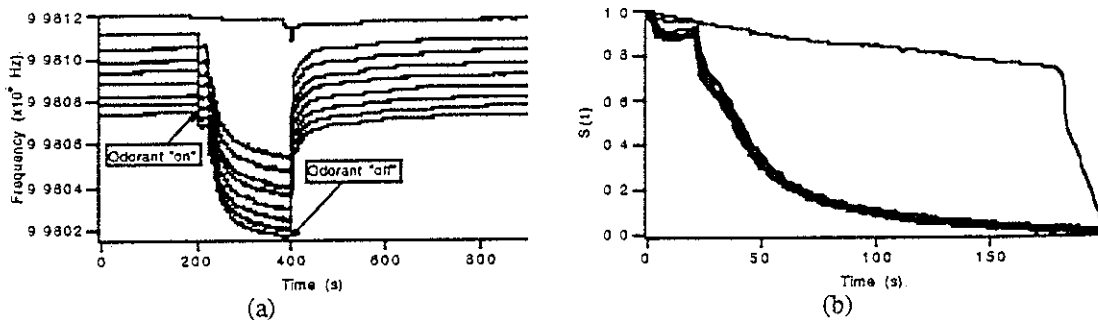


Figure 2. (a) The frequency response of an OV-17 coated sensor to 25 separate exposures of an ethyl acetate odorant. For clarity only every third exposure is shown. (b) The corresponding kinetic signatures, $S(t)$ of the full 25 exposures.

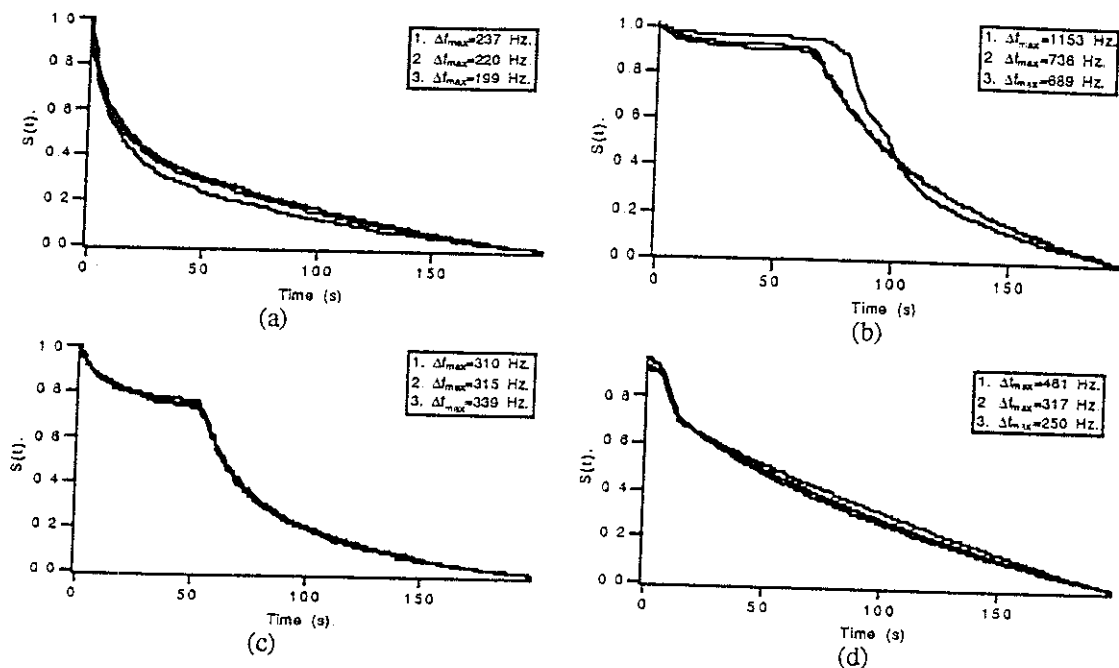


Figure 3. Sample kinetic signatures and corresponding peak frequency response of various odorant-sensor combinations: (a) Dicyclohexylamine, (b) piperidine and (c) n-methyl piperidine odorants on pyridoxine hydrochloride + antarox CO-880 coated sensor (d) Napthalene odor on ascorbic acid coated sensor.

ascorbic acid and OV-17, this latter coating being used previously as the basis of a camphor-guided robot [6]. The sixth sensor was an uncoated (clean) crystal

Experiments were carried out in a gas flow chamber [10]. Briefly this creates a constant flow (200 ml min^{-1}) across the sensor of dry deodorised air. During odorant stimulus the odorant stream is introduced perpendicular to the sensor surface, also at 200 ml min^{-1} to minimise the effect of pressure fluctuations when switching between the two sources of flow. The change in resonant frequency of the sensor is monitored and the kinetic signatures of the six sensors is presented to the ANN for analysis.

Sample kinetic signatures from the range of responses obtained, and their corresponding peak frequency responses are shown in figure 3. The comparative merits of the two forms of analysis (kinetic signature vs peak frequency response), are further demonstrated; the kinetic signature responses show a high degree of repeatability. Contrast this with the peak frequency responses of the same sensors (inset), and it becomes obvious that there is a high degree of variability in succeeding exposures; the measured difference in the peak frequency of the third exposure in (b) and (d) being 60% and 52% respectively of the original response. Clearly kinetic signature analysis offer a

more consistent means of representing the odor-sensor interaction

4. An electronic nose for detecting odorants.

The recognition of odorants using kinetic signature analysis requires a system to correctly classify odorants with the corresponding kinetic signatures generated from multiple sensors. The ANN that was constructed to perform this mapping is illustrated in figure 4.

Processing of kinetic signatures is performed by tiered, three layer back propagation ANNs, with each tier representing the dynamic response of each of the sensors ($i=1,2,\dots,6$) that comprise the electronic nose. Input to the system during learning and recall is via a matrix $X_j^k(i,r)$ for each presentation k , of an odorant ($j=1,2,\dots,18$) to the 'nose'. Here the X matrix is defined as the response of all the sensors i , to consecutive k exposures of the j^{th} odorant. The matrix is comprised of a series of rows (one for each sensor), whose elements r , are derived from a vector $T_{ij}(r)$ that characterise the response of each sensor i to an odorant j . Output of the ANN is a weighted, two dimensional output matrix $O^k(i,j)$, constructed from the output processing elements (PEs) of the

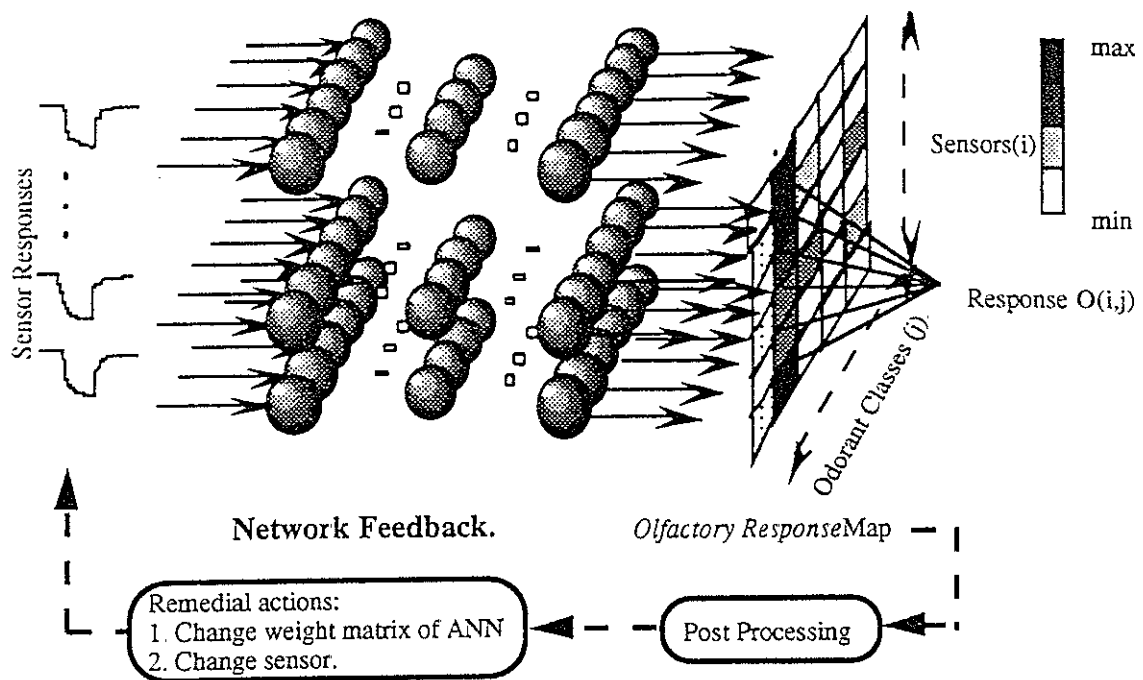


Figure 4. Schematic representation of the electronic nose, illustrating the functional units of the system: Using simple post processing on the response map the system can detect when either i) sensors have been damaged, or ii) an unknown odorant has been encountered. Possible solutions to these problems are to either modify the weight matrix of the ANN to accommodate new sensor responses (impractical using current configuration), or alternatively replace the damaged sensor(s).

ANN to give the *olfactory response map* (see figure 5)

The vector $T_{ij}(t) = (r_1, r_2, \dots, r_{66})$ incorporates a measure from three of the most important variables observed: i) Nine variables containing a representation of the peak frequency response of the sensor-odorant interaction, encoded in a field delimited by specific frequencies: {(Hz): 10,000: 1,000: 700: 300: 100: 50: 20: 10: 1}. For example a peak frequency shift of 374.2 Hz would be encoded as [0,0,0,1,0,1,1,0,4,0,2]. Representation of values within the peak frequency field is based on experimental observation, and are selected to give maximum impetus within the network to lower frequency responses, as it is within these regions that instrument noise can impair the resolution of the kinetic signature. ii) A compressed version (to speed learning) of the kinetic signature $S(t) = (t_1, t_2, t_3, \dots, t_{197})$. iii) Lastly, seven variables are appended to the vector to convey information about the sensors history of response to the odorant. They are encoded in a field delimited by the values [1, 2, 3, 5, 10, 30, 100], so a sensor that had registered a history of 4 successive exposures to the odorant would be represented by [1,0,1,0,0,0,0]. This format is weighted towards the lowest number of exposures as we have found that the most pronounced changes in kinetic signatures occur in the initial reactions between odorant and sensor.

Training of the ANN is by repeated presentations ($n=2000$) of the learning matrix $X_j^k(i,r)$ to the ANN. Learning is enacted using the back propagation learning algorithm [14] concurrently over each separate tier of the ANN. Separate learning rate parameters $\alpha=0.30$ and $\beta=0.15$ are used to control the rates of weight change during learning in the hidden and output PEs respectively. Construction of each tier of the ANN was a standard three-layer back propagation ANN: The activation function for each PE was the logistic sigmoid ($f(x)=1/(1+e^{-x})$), and dimensions of the amalgamated ANN were 6 tiers \times (66 input layer PEs + 30 hidden layer PEs + 18 output layer PEs (one for each odor class)) = 684 PEs.

To evaluate how well the trained ANN discriminates between the odorants these patterns were again presented to the ANN. The response is illustrated in table I. The first figure is the accuracy of response A_j , of the ANN for the incident odorant j . This is defined to be the mean response elicited for each of the sensors ($i=1,2, \dots, s$) to p independent presentations of the odorant j , to the ANN.

$$A_j = \frac{\sum_{k=1}^p \sum_{i=1}^s O^k(i,j)}{ps} \quad (2)$$

<u>Odorant</u>	<u>Accuracy of Response</u>	<u>Confusing Odorant</u>	<u>Response</u>
1 - Diethylamine	(94.75%)	<i>Octyl Acetate</i>	(1.46%)
2 - Triethylamine	(92.54%)	<i>Diisopropylamine</i>	(2.43%)
3 - Isopropylamine	(87.21%)	<i>Diisopropylamine</i>	(7.15%)
4 - Diisopropylamine	(75.27%)	<i>Isopropylamine</i>	(7.25%)
5 - Diisopropylethylamine	(95.32%)	<i>Camphor</i>	(1.80%)
6 - Dicyclohexylamine	(90.23%)	<i>Hexyl Acetate</i>	(1.08%)
7 - Piperidine	(95.50%)	<i>Triethylamine</i>	(1.07%)
8 - n-methyl Piperidine	(96.17%)	<i>Diisopropylamine</i>	(1.55%)
9 - Citronellol	(95.08%)	<i>Isopropylamine</i>	(1.23%)
10 - Camphor	(94.88%)	<i>Diisopropylethylamine</i>	(2.11%)
11 - Naphthalene	(94.75%)	<i>Camphor</i>	(1.93%)
12 - Methyl Acetate	(86.21%)	<i>Triethylamine</i>	(4.80%)
13 - Ethyl Acetate	(89.60%)	<i>Propyl Acetate</i>	(2.20%)
14 - Propyl Acetate	(94.27%)	<i>Ethyl Acetate</i>	(1.28%)
15 - Amyl Acetate	(94.92%)	<i>Hexyl Acetate</i>	(1.55%)
16 - Hexyl Acetate	(94.48%)	<i>Methyl Acetate</i>	(1.22%)
17 - Heptyl Acetate	(85.31%)	<i>Octyl Acetate</i>	(7.54%)
18 - Octyl Acetate	(88.96%)	<i>Heptyl Acetate</i>	(6.13%)

Table I. Accuracy of response of the trained ANN.

The figure in italics represents the next highest response from any of the other odorant identifying output PEs (i.e. it indicates the level of confusion). These responses are all generally less than 5%, demonstrating that the ANN shows excellent ability in discriminating between chemically similar odorants. A few of the odorants do show notable confusion; these are di/isopropylamine and octyl/heptyl acetate. This similarity in response is most probably due to contamination by their pairs (i.e. in diisopropylamine vapour there will be a small concentration of isopropylamine molecules present and vice-versa).

The illumination of these chemical associations as illustrated in table I, give chemical credence to the use of kinetic signatures for

characterisation of odorants using QPZ sensors. The figures also suggest that there may be potential use for this type of analysis as a quick method of determining the analytical purity of chemical solutions using vapour analysis.

5. System feedback using the olfactory response map.

The environment in which a chemically based sensor is to operate will in most cases be hostile, as there is no way of determining how individual sensors will react to the thousands of different gaseous molecules they might encounter. A durable electronic nose must therefore be capable of detecting and adapting to changes in sensor responses immediately after they occur. In this

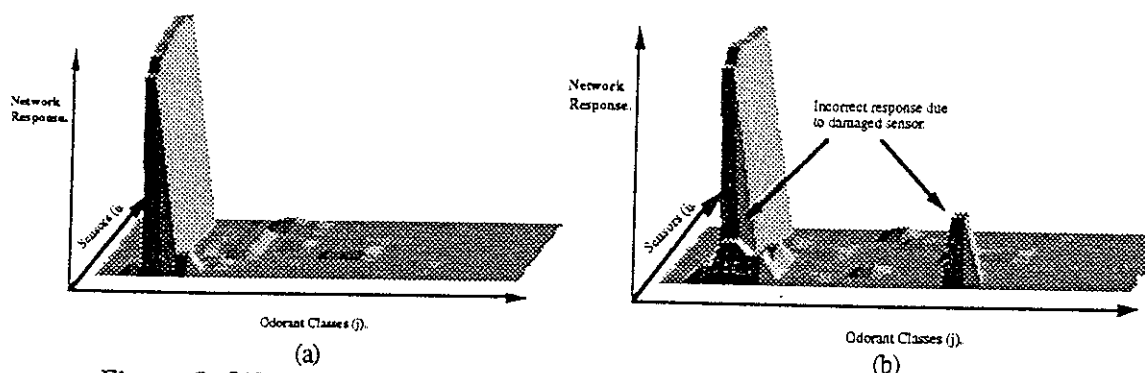


Figure 5. Olfactory response maps, generated for the first (a) and third (b) exposure of the odorant isopropylamine to the electronic nose. Note in the third exposure, an errant response is recorded for one of the sensors ($i=1$). This is most probably due to a marked chemical change on the sensors surface in the proceeding two exposures, causing its response to the odorant to change dramatically.

section, we discuss how the ANN as depicted in figure 4 can be used by an automated system to adapt its physical and/or interpretive state to maintain the integrity of the electronic nose in response to modification of sensors by non-specific gaseous molecules

Classification of odorants could have been performed with similar clarity by connecting the outputs of the hidden layer PEs to a single tier of output PEs, with each PE (1,2,...,j) representing the ANNs identifying response to each of the odorants j. The matrix response was chosen, because it presents a visible means of monitoring individual sensor response, and also avails itself to higher order processing (e.g smoothing, filtering, ...) that may be further used to enhance the quality of odorant identification.

The *olfactory response map* $O^k(i,j)$ for the odorant isopropylamine is shown in figure 5, for the first ($k=1$) and third ($k=3$) presentations of the odorant to the 'nose'. Note the incorrect response of sensor 1 in figure 5(b) to the odorant, which registers notable confusion with another of the odorant classes, as is indicated by a sharp peak to the right. The cause is most probably due to marked chemical changes occurring at the surface of the sensor during previous exposure, and the ANN fails to resolve this new response with the correct odorant class. Note, however that by simple summation of the response $O(i,j)$ for each sensor i , the correct odorant is identified.

It was mentioned earlier that variability in sensor response to repeated odorant exposure is a prevalent problem in gas sensing due to the unknown volatility of the sensor coating and odorant interaction. While kinetic signatures offer a far more repeatable means of representing odorant-sensor interaction; the response of the sensor can still be altered dramatically by highly reactive gases, or the cumulative effect of adsorption over time. In the absence of a suitable remedial mechanism, the response of the ANN will eventually become degraded and its ability to discriminate between odors compromised.

This problem is similar to that encountered by the mammalian nose. In gas sensors, the odorant-receptive medium is the chemical applied to the surface of the sensor. The mammalian equivalent is protein receptors found on cilia projecting from dendrites of olfactory sensory neurons. These dendrites project to the periphery of the nasal cavity protected only by a thin layer of mucus, and their proximity to the external environment makes them highly susceptible to infection by airborne viruses and chemical trauma. To maintain the integrity of the olfactory system, olfactory sensory neurons must therefore be capable of regeneration throughout much of the lifespan of

an individual [9]. The incorporation of an equivalent capability into an electronic system is eminently desirable, as it would effectively nullify the destructive effect airborne volatiles can have on the response of the sensors.

The ability to recognise sensor damage is therefore one of the principle benefits of organising the response of the ANN into these *olfactory response maps*. An automated system could test for sensor damage, using a simple filter that identifies a particular sensor which is not responding to a odorant, as signified by the responses of the sensors around it (e.g. figure 5(b)).

Following detection of a damaged sensor there are two possible courses of action for an automated system i) The sensor is replaced automatically and the nose registers where and when the errant event occurred. ii) Alternatively a more practical method would be to alter the weight matrix of the ANN to accommodate the 'new' response of the sensor to the odorant. This is of course not feasible in the current configuration given the inflexibility of the back propagation learning algorithm, and a more plastic ANN classifier such as ART2 [4], might be more appropriate in this instance.

Regardless of the technique employed, the intrinsic value of this form of feedback is to offer an automated system an additional level of computation, allowing it to modify its internal state in response to changes caused by external factors.

6. Discussion.

We have demonstrated the preliminary framework of an improved electronic nose that incorporates i) in kinetic signatures; a reproducible means of representing odorant-sensor interactions and ii) a novel ANN configuration that offers a remedial mechanism well suited for operation in volatile environments. These features lay the groundwork for the development of more dynamic electronic olfactory systems, capable of operating autonomously in diverse odorous environments.

The instrument [10] on which trials were carried out was initially designed to monitor the dynamic response of QPZ sensors under differing experimental conditions. The peripheral equipment designed to facilitate this analysis make it too bulky for field use, and a portable version that will speed up sensor response times is currently under construction.

As physical replacement of damaged sensors is a awkward feature to implement in a robotic system, the practice of retraining the internal ANN to accommodate sensor damage is the preferred option. To better facilitate this feature an

ANN is currently being developed. It will be highly plastic, have an 'early-out' option when processing kinetic signatures so as to speed up recognition times and be capable of handling unknown odorants (odorants that are not part of the training set)

The enormous potential offered by electronic olfactory systems has not yet been met by current technologies. In part this is due to the practice of applying rigid mathematical structures for the analysis of what is, not unlike the mammalian nose a very dynamic system. An appreciation of the volatile nature of the medium in which an electronic nose must operate, and correspondingly more dynamic forms of analysis such as those demonstrated, will inevitably lead to the development of more sophisticated electronic noses in the future.

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