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# An electronic nose based on solid state sensor arrays for low-cost indoor air quality monitoring applications

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#### Abstract

The occurrence of illnesses related with poor ventilation has driven an increasing attention towards indoor air quality monitoring. In buildings equipped with climate control systems, the diseases related to the air quality can be significantly reduced if smart intervening procedures, aiming to control the concentration of pollutants in the indoor air, can be implemented in the heating, ventilation air conditioning unit. When reliable information about both the indoor and outdoor air quality is made available, the climate control system can provide the most appropriate amount of ventilation, ensuring safe and comfortable living conditions.

In this paper, a dedicated, miniaturized, low-cost electronic nose based on state-of-the-art metal oxide sensors and signal processing techniques was developed. The proposed device is targeted to the quantification of carbon monoxide and nitrogen dioxide in mixtures with relative humidity and volatile organic compounds by using an optimized gas sensor array and highly effective pattern recognition techniques. The electronic nose was tested in an environment reproducing real operating conditions. Exploiting the unique response patterns of the different sensors in the array and the capability of a simple fuzzy-logic system it was possible to identify and discriminate concentrations as low as 20 ppb for NO<sub>2</sub> and 5 ppm for CO in the test gas environment, allowing to reach the necessary sensitivity towards the target pollutants together with the selectivity towards the typical interfering gas species.

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# 1. Introduction

The increasing interest in indoor air quality (IAQ) monitoring is mainly due to the growing incidence of a new class of diseases, identified as building-related illnesses (BRI) and sick building syndromes (SBS), arising from long-term occupancy of confined living spaces, like office buildings or homes/apartments. The occurrence of these diseases is related with the presence of physical, biological and/or chemical contaminants inside the building. The effects on the human health due to indoor pollution include pathologies for which the etiologic agent has been precisely identified and pathologies not directly linked with a specific physical, chemical or biological specie, since the symptoms arise from the concomitant effect of several pollutants which are all present in the ambient, e.g. from low air quality [1]. Chemical contaminants, and in particular gaseous compounds, were found to be among the main responsible ones for both the BRI and the SBS diseases, since in buildings with inadequate ventilation they can accumulate with time.

Typically, we can find several hundreds of indoor chemical contaminants, including by-products of the combustion (NO<sub>2</sub>, SO<sub>2</sub>, CO, etc.), cigarette smoke, particulate matter, mineral fibers and a number of volatile organic compounds. In spite of the very low concentrations, some of these compounds are extremely toxic, like NO<sub>2</sub> or CO; some other, like benzene and formaldehyde, were proved to be carcinogenic. Therefore, the monitoring of the air quality is of paramount importance to keep safe and healthy conditions.

In buildings equipped with heating ventilation air conditioning (HVAC) systems, the pollutants are diluted by ventilation, which is generally operated on the basis of fixed duty cycles. This approach does not necessarily ensure an improvement of the indoor air quality, since in many cities "fresh" outdoor air can contain many pollutant species at concentrations higher than the threshold values. Furthermore, an uncontrolled preventive increase of the ventilation results in an increment of the overall energy consumption,

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Table 1 Typical indoor air quality contaminants

<b>9</b> I	1 2	
Aldehydes		
Ozone		
Nitrogen dioxide		
Carbon dioxide		
Particulate matter		
Radon		
Carbon monoxide		
Sulfur dioxide		
Total VOC		
Formaldehyde		
Water vapor		
Lead		

especially when the temperature gap between indoor and outdoor air is non-negligible [2].

Several attempts to implement demand-controlled ventilation have been reported in literature. Many of them rely on the quantification of CO<sub>2</sub>, used as a tracer of human occupancy in confined living spaces or as surrogate of inhabitant generated pollution. However, this approach is inadequate to monitor IAQ, since several toxic compounds released by building materials and furnishings or generated by human activities are also present in the indoor environment. Since each compound has a different impact on human health, it is important to monitor their concentrations individually. Table 1 reports a list of the most common indoor contaminants found in confined living spaces, which can be used as tracers for the IAQ [1,3,4].

For this reason, the availability of reliable, low-cost sensors suitable to monitor both the indoor and outdoor air quality would allow the implementation of smart HVAC intervening procedures, considering not only the typical information about temperature and relative humidity but also the concentration of a number of compounds used as air quality tracers. This way, by driving the ventilation systems on demand to keep the IAQ under control, it would be possible to maintain acceptable healthy and comfortable conditions while minimizing the overall power consumption. Being the information about the quality, the humidity and the temperature available for the inside and the outside air, the climate control system would intake fresh air, recycle the indoor air or operate active scrubbers, depending on the most favorable energetic conditions.

Some low-cost tools for IAQ analyses are nowadays commercially available, but they are not suitable to provide reliable information, since the various pollutants are not precisely identified and quantified. These devices usually give indications about the overall IAQ, without any estimation of the concentration of each pollutant. In fact, only the detailed information about the concentration of the single gas would allow an optimized control of the HVAC system, including the activation of scrubbers to catalytically convert some pollutants, like, e.g. volatile organic compounds, in  $CO_2$  and water vapor.

In this work, an electronic nose (e-nose) based on a solid state gas sensor array for the identification and the quantification of two typical indoor air quality tracers is presented. The simplicity of the proposed approach, which uses stand-alone gas sensors, simple driving electronics and fuzzy-logic pattern recognition algorithms, aims to realize a low-cost tool suitable to monitor some of the compounds of interest for IAQ. The availability of this type of devices is fundamental for a capillary analysis of the pollution level inside buildings equipped with forced ventilation and air climate control units. To validate the approach, only CO and NO<sub>2</sub> were taken into consideration as target compounds, while some VOCs and the humidity were considered as interfering species.

# 2. Experimental

The aim of this work is to develop a reliable tool for IAQ monitoring using a very simple system architecture based on a stand-alone gas sensor array, driving electronics and suitable pattern recognition algorithms, without any fluidic components (valves, filters, pumps, etc.). Furthermore, the system has been designed to integrate additional sub-units able to monitor other parameters relevant for IAQ. To validate this approach, the number of compounds considered within this work was limited to two gases chosen among those reliably detectable by metal oxide solid state gas sensors. Table 2 reports these gases together with the target concentration ranges considered for effective IAQ monitoring and the relevant indoor threshold values as defined in [3]. In spite of their relevance for indoor comfort, some other compounds, like carbon dioxide  $(CO_2)$ , were not considered here because their detection is problematic with metal oxide gas sensors. However, other sensing devices capable to detect these compounds are nowadays commercially available and they could be easily integrated into a modular architecture.

Table 2

Some gas species considered as IAQ tracers and their target concentration ranges

Compound	Concentration range	Indoor threshold values (8 h exposure)
СО	5–30 ppm	9 ppm
NO <sub>2</sub>	20–200 ppb	53 ppb <sup>a</sup>
VOC (benzene, toluene, <i>m</i> -xylene)	60–600 ppb	n.a. <sup>b</sup>
Water vapor (RH)	5–95%	No threshold

<sup>a</sup> US-EPA suggested value for 24 h exposure outdoor threshold. No indications given for indoor exposure.

<sup>b</sup> No standards have been set for VOCs in non-industrial indoor settings

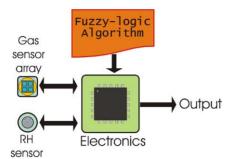


Fig. 1. Schematic overview of the considered e-nose architecture.

The system developed for this work consists of an array of thin-film metal-oxide gas sensors, driven by ad hoc electronic circuits suitable to power and measure each device independently. The gas sensors were operated under different conditions and they provided distinct response patterns. A commercial RH sensor monitored the relative humidity, which interferes in both CO and NO<sub>2</sub> detection. The discrimination between the different target gases and their quantification were achieved using state-of-the-art data processing and pattern recognition algorithms, which rely on the distinct response patterns provided by the sensors. In fact, the low selectivity of the solid state gas sensors has been compensated by the combination of the response patterns, allowing the quantification of the single gas species. In this work, a fuzzy-logic system was used for pattern recognition. A similar approach, based on a multi-sensor array and using an artificial neural net (ANN) for pattern recognition, has been proposed in [5].

A schematic drawing of the unit is shown in Fig. 1. This simple architecture is targeted for the detection and quantification of CO and NO<sub>2</sub>, but it does not allow a precise quantification of volatile organic compounds (VOC), because of the lack of selectivity in metal oxide semiconductor gas sensors. We are currently addressing this problem following an approach based on a gas-chromatographic architecture realized by MEMS technologies and developed for the detection and quantification of VOC [6], which will be described in a future paper. Both the dedicated gas-chromatographic architecture and the simple e-nose presented in this work are conceived to be part of a modular system for innovative IAQ monitoring applications.

## 2.1. Metal oxide resistive gas sensors

The solid state gas sensors used for the detection of the air quality tracers are based on thin films of metal oxide semiconductor materials [7,8] deposited onto state-of-the-art micromachined hotplate arrays integrated in a single 5 mm × 5 mm Si chip [9]. These hotplates consist of four suspended 200 nm thick Si<sub>3</sub>N<sub>4</sub> membranes integrating a buried platinum heater and passivated by a 800 nm thick SiO<sub>2</sub> layer. The very low thermal mass of the suspended structure allows to reduce the power consumption with respect to commercial devices (only 60 mW to operate the sensor at 400 °C), as well as to heat up and cool down the sensing element very fast, which is necessary for fast pulsed temperature (FPT) mode sensor operation [10].

On top of these hotplates, we deposited two types of gas sensing materials by means of different deposition techniques, namely tin oxide  $(SnO_2)$  by rheotaxial growth and thermal oxidation (RGTO) [7] and tungsten trioxide (WO<sub>3</sub>) by pulsed laser ablation (PLA) [8]. The morphology of the thin films achieved through the optimization of the deposition techniques leads to very high sensitivity towards the considered gas species, allowing the detection of the gas concentrations down to the range required for IAQ.

The lack of selectivity of resistive gas sensors was partially compensated by the deposition of catalyst materials as well as very thin SiO<sub>2</sub> caps above the sensing layer. The response towards the single gas species was further enhanced through the use of different operating temperatures and by operating the sensors in FPT mode [10]. Table 3 shows some of the optimal combinations of materials and operating conditions and the corresponding target gas species. The optimization of both the sensing material and its operating conditions results in excellent sensor responses towards CO and NO<sub>2</sub>. On the other hand, the low selectivity of metal oxide semiconductor gas sensors was only partially overcome by the material optimization, since interference phenomena by water vapor and VOC still affected the detection, as will be discussed later.

## 2.2. Characterization protocol

The characterization was performed with a specifically designed characterization system, where up to eight pollutants could be simultaneously injected into a Pyrex measurement chamber. The large inner volume (321) of the chamber and the applied flow rate (500 sccm) were chosen to replicate as close as possible the operating conditions found in a room equipped with a HVAC system, where every hour a complete room volume of air is pumped-in. During the test, mixtures of all pollutants at three different concentrations were injected into the characterization chamber. The

Table 3 Sensing materials and operating conditions used to detect the different gas species

Sensing layer material	Catalyst	Operation mode	Operating temperature (°C)	Target gas species	
SnO <sub>2</sub>	Au	FPT	375	NO <sub>2</sub>	
WO <sub>3</sub>	Au	DC	250	NO <sub>2</sub>	
$SnO_2 + SiO_2$	Au	DC	400	CO	
SnO <sub>2</sub>	_	FPT	325	CO	

gas distribution system delivers synthetic air, humidified air and the target pollutants from certified gas cylinders in balanced quantities to get the final concentration of each compound. The system is completely controlled by a PC running a specifically developed software package, which allows the recursive execution of automatic sequences lasting several weeks. The sensor arrays are driven and controlled by a custom-made electronic interface connected to a further PC, which allows the simultaneous and completely independent acquisition of up to 16 sensors. The sensing layer resistivity, i.e. the sensor response, was sampled by applying a 1.2 V bias to the sensing layer and measuring the flowing current. Finally, a standard GC/MS is connected to the measurement chamber, performing spot-wise quality control tests of the gas mixture inside the chamber.

The set of data used for the analysis presented in this work consisted of 2000 patterns acquired within the last 45 days of a 60-day sensor characterization run. The data relative to the first 15 days were neglected, since the sensors used in this characterization were brand-new and had undergone no stabilizing burn-in procedures, thus showing a considerable drift of the conductivity during the first operation period.

## 2.3. Pattern recognition techniques

The characterization data specifically collected during long-term measurement campaigns lasting more than 2 months, were then used to train and test a fuzzy system, which provided an estimation of the initial concentration of each target compound in the gas mixture.

Operating the sensors under optimized conditions for the detection of a given compound does not make them fully selective, but they do provide unique response patterns, which can be used to classify and quantify the single pollutants inside complex gas mixtures, as found in real applications. To evaluate concentration of each compound in a complex gas mixture, we used a neuro-fuzzy system provided by the adaptive fuzzy modeler (AFM) developed by ST-Microelectronics [11]. AFM is a software tool for the automatic fuzzy model generation of a system starting from its input-output data sampling. It implements neural training algorithms for the successive optimization of both the rules and the sets of a simple Sugeno-type zero-order fuzzy system. The automatic rules identification is carried out, thanks to an unsupervised learning on a winner-take-all fuzzy associative memory neural network, while the optimization of the fuzzy sets parameters is carried out by a supervised learning on a multilayer backward-propagation fuzzy associative memory neural network [11]. As fuzzy intersection operators (inference methods), AFM allows to select either the minimum or the product operator. The AFM package is supplied for feasibility studies of ST-Microelectronics' fuzzy-logic ASICS (W.A.R.P. 1+2, ST52x301), which implement all the processing capabilities of the fuzzy systems simulated in the AFM. Therefore, the processing electronics necessary for the realization of the e-noses studied in this work are already commercially available. Further details on fuzzy logic can be found in [12].

The AFM was used to simulate two fuzzy systems having three input variables and one output prediction each, being the input variables the output from the CO, NO<sub>2</sub> and RH sensors and the output predictions, the CO and the NO<sub>2</sub> concentrations, respectively. In the case of the CO sensor, five Gaussian fuzzy sets were used for the CO input and three Gaussian sets for each of the interfering NO<sub>2</sub> and RH inputs. In the case of the NO<sub>2</sub> sensor, five Gaussian fuzzy sets were used for the NO<sub>2</sub> input and three Gaussian sets for each of the interfering CO and RH inputs. The system was trained with the product inference operator for 50 learning epochs and converged rapidly.

#### 3. Results and discussion

### 3.1. Response of the gas sensor arrays

The optimization of the sensing layer materials and of the operating conditions has allowed to reach the sensitivity necessary for the reliable detection of CO down to concentrations of 5 ppm and of NO<sub>2</sub> down to 20 ppb. To detect CO, we used a SnO<sub>2</sub> + SiO<sub>2</sub> sensor with Au catalyst operated in DC mode at 400 °C, while the sensor targeted for NO<sub>2</sub> was a SnO<sub>2</sub> sensor with Au catalyst operated in FPT mode with a pulse temperature of 375 °C, a duration of 1 s, and a period of 60 s. Other sensors were used during the extensive characterization run, principally FPT mode operated SnO<sub>2</sub> sensors for CO detection and WO<sub>3</sub>-based sensors for NO<sub>2</sub> detection, but the validation of the sensor response patterns during the training of the fuzzy system have shown that these combinations are less effective, and therefore only the sensor responses of the first two sensor types will be considered.

Fig. 2a and b shows the typical response of two sensors targeted for the detection of CO and NO<sub>2</sub> to three different concentrations of these pollutants. First, the sensor baseline in synthetic air with RH = 50% is shown, up to T = 0 min. Afterwards, three different concentrations of CO or NO<sub>2</sub> are consecutively injected and kept each one for 15 min. In this case, the considered concentrations were 5, 15 and 30 ppm for CO and 20, 100 and 200 ppb for NO<sub>2</sub>. The plots of Fig. 2 emphasize the excellent response of the sensors. For the CO sensor, the increase of the conductivity for the first concentration of 5 ppm amounts to approximately 400%, while the NO<sub>2</sub> sensor shows the typical effect of an oxidizing gas specie on n-type semiconducting materials, with a variation of the resistivity of approximately 300% for the 20 ppb concentration. While the CO sensor shows a good dynamic response over the entire concentration range, the NO<sub>2</sub> sensor saturates at the higher concentrations ( $[NO_2] > 100 \text{ ppb}$ ). However, in the range of the indoor threshold value  $[NO_2] =$ 53 ppb, the sensor features good discrimination capabilities. From these results it is important to evidence that, having response variations three to four times the baseline value,

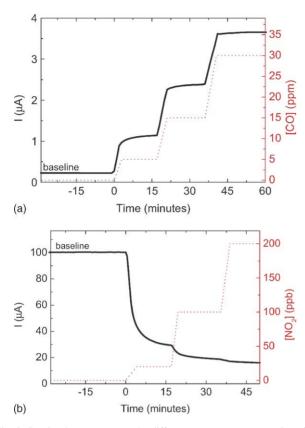


Fig. 2. Sensing layer current at the different target gas concentrations for the CO sensor (a) and the NO<sub>2</sub> sensor (b) at RH = 50%.

the detection limit of our sensors for both CO and  $NO_2$  is well below the lowest concentration considered in this work, ensuring a high degree of confidence for the concentration predicted by the fuzzy system.

The plots in Fig. 2 do not give information about the selectivity of the sensors, since the responses shown are relative to the injections of a single pollutant at fixed RH values. Furthermore, only a single measurement shift is shown, and the possible drift of the baseline with time cannot be disclosed. To evaluate the overall sensor selectivity and the evolution of the device response with time, we compared the conductance of the sensing layer element of both the CO and NO<sub>2</sub> sensors for the different concentrations of the target pollutant with and without the other gases in the mixtures. The results for a 45-day measurement period are shown in Fig. 3a and b. More in detail, Fig. 3a shows the currents of the CO sensor when 0, 5, 15 and 30 ppm of CO are present in the measurement chamber, in any mixture with NO<sub>2</sub> and VOC at different concentrations. For simplicity, only the measurements acquired with RH = 30% are shown. Conversely, Fig. 3b shows the currents of the NO<sub>2</sub> sensor when 0, 20, 100 and 200 ppb of NO<sub>2</sub> are injected with RH = 30%, in any mixture with CO and VOC. From Fig. 3 it can be pointed out that, during 45 days of operation, the sensors exhibit a good sensitivity towards the considered gas species together with a certain sensor drift with time. Fig. 3 also show a spread of the single measurements, which is re-

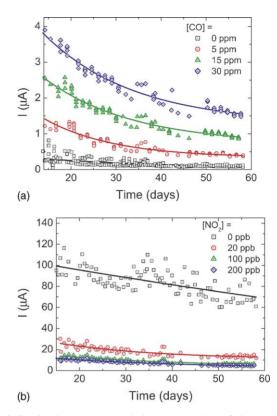


Fig. 3. Sensing layer current evolution over 45 days of characterization at the different concentrations of the target gases in mixtures with all interfering species, for the CO sensor (a) and the  $NO_2$  sensor (b).

lated with some cross-response to the other interfering compounds present in the gas mixture. A detailed analysis of the sensor response shows that especially the relative humidity and the NO<sub>2</sub> concentration interfere in the CO detection, while the NO<sub>2</sub> sensor is affected primarily by interference from humidity, only. As far as the VOCs are concerned, only very small interference on the CO sensor can be observed, but this cross-sensitivity can be neglected if compared to the excellent sensor response towards CO. In fact, the optimized conditions for VOC detection were found to be quite different from those used for CO and NO<sub>2</sub> detection.

These results imply that the selectivity exhibited by our sensors is still not enough to allow for a direct use of the sensor output for the quantification of the gas concentration. However, being the information acquired by a commercial RH sensor included as input to the fuzzy system, the combination of the input patterns from the three sensors should allow for the extraction of the concentrations of the two target gases, CO and NO<sub>2</sub>.

Furthermore, if we consider the sensor response, defined as  $(G_{GAS} - G_0)/G_0$  or  $(R_{GAS} - R_0)/R_0$  for oxidizing and reducing gas species, respectively, this parameter is quite stable for both the CO and the NO<sub>2</sub> sensors, but the overall sensor conductivity sensibly decreases with time. In a real application, direct information about the baseline conductivity is not available unless a calibration or an air purifier unit has been integrated into the system. If the target is a

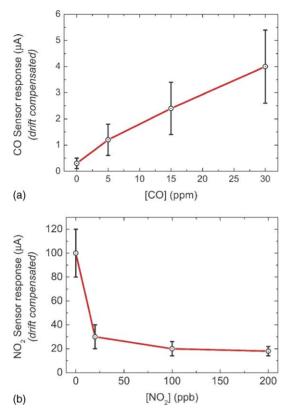


Fig. 4. Pre-processed sensor response spread as a function of the target gas concentration, prior to fuzzy-logic pattern recognition, for the CO sensor (a) and the NO<sub>2</sub> sensor (b). The error bars are relative to the  $2\sigma$  interval.

low-cost e-nose, the drift of the sensor conductivity with time has to be analytically compensated before supplying the sensor data to the fuzzy pattern recognition system, and this is easily possible due to the smooth exponential decay of the conductivity with the time, which can be disclosed from Fig. 3.

However, the baseline drift compensation remains a critical issue, since the nature of the conductivity decay could not be completely explained yet. In this concern, investigations about the nature of the drift are in progress, and first results show a good reproducibility of the conductivity decay shape.

Fig. 4a and b resumes the quantification capability of the single gas sensors, prior to the application of the fuzzy-logic pattern recognition algorithms. The plots show the raw characteristics of the single sensor's currents as a function of the target gas concentration, for all the measurements acquired within the 45 days of characterization. Fig. 4a is relative to the CO sensor, Fig. 4b to the NO<sub>2</sub> sensor, and the data is already being compensated for the exponential drift in the sensor current.

From the plots in Fig. 4, one can see that the sensors are partially selective and show a clear trend of the sensing layer current with the target gas concentration. However, the simple response of the single sensors is not sufficient for the precise quantification of the pollutants. The error bars reported

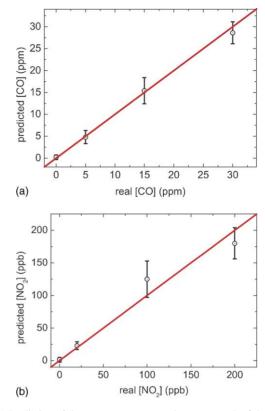


Fig. 5. Prediction of the target gas concentrations as a result of the multisensor response pattern combination performed by the fuzzy-logic algorithm: CO prediction (a) and NO<sub>2</sub> prediction (b). The error bars are relative to the  $2\sigma$  interval.

in the plots represent the two standard deviation  $(2\sigma)$  interval of all the sensor output values at a fixed concentration. As shown in the plots, the error bars relative to the single concentrations overlap with the neighboring concentrations, due to the cross-response to the interfering gas species. For this reason, a pattern recognition technique must be applied, in order to combine the responses of the different sensors, allowing the prediction of the final target gas concentrations.

## 3.2. Fuzzy pattern recognition algorithm

The plots in Fig. 5a and b show the results of the prediction for both the CO and the NO<sub>2</sub> concentrations obtained applying the two fuzzy systems to the collected set of data. From the 2000 response patterns, 1400 were randomly extracted and used for the training of the fuzzy system and the remaining 600 were used for the validation of the recognition algorithm. The error bars are relative to the  $2\sigma$  interval of the various predictions for each concentration.

As can be seen from these figures, considering the simplicity of the proposed approach, the estimated values of the concentrations are in excellent agreement with the real pollutant concentrations injected into the mixture. Using this approach, the three concentrations for CO can be easily discriminated, while for NO<sub>2</sub> only the highest concentrations may overlap. This behavior can be explained by considering Fig. 2b, which shows the excellent sensor response towards the low concentrations and the significant saturation effects between the 100 and 200 ppb concentrations. This means that the NO<sub>2</sub> sensor is that sensitive to already saturate at these sub-ppm concentrations. This problem can be easily overcome by applying slightly different operating conditions for the NO<sub>2</sub> sensor, such as a higher operating temperature. This way the sensitivity at low concentrations decreases, but the slope of the response at concentrations higher then hundred ppb become more pronounced, allowing for better separation and discrimination between each concentration. However, in our application we focused more to the range below 100 ppb, where concentrations of NO<sub>2</sub> as low as 20 ppb can be very well identified and separated.

## 4. Conclusions

In this work, a simple e-nose architecture for indoor CO and NO<sub>2</sub> monitoring is presented. This instrument belongs to a new class of low-cost e-noses for air quality monitoring to be integrated within the climate control unit, which would allow a more efficient use of HVAC systems, keeping the IAQ under control while lowering the overall power consumption.

The e-nose is based on an array of solid state metal oxide semiconductor gas sensors realized onto micromachined hotplate arrays. The use of a micromachined substrate, the optimization of the deposition techniques as well as the use of the most appropriate operating parameters, allowed for the realization of low-cost, low power consumption devices with high sensitivity and enhanced selectivity.

These devices were tested in an environment reproducing real operating conditions and concentrations as low as 20 ppb of NO<sub>2</sub> and 5 ppm of CO were continuously monitored for more than 45 days of extensive sensor characterization, which showed no significant degradation in the sensor response. The target gas concentrations in the mixtures have been precisely estimated exploiting the capability of a simple fuzzy-logic system used to combine the unique response patterns of the various sensing elements and to extract the most significant information from the sensor data. Using this approach, it was possible to identify and discriminate the presence of each pollutant and to estimate the composition of the air mixture inside the test environment. The presented results show the feasibility of low-cost e-noses developed to detect CO and NO<sub>2</sub> at concentrations lower than the IAQ threshold values, suitable for the integration in HVAC systems.

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