Measurement requirements for environmental monitoring : application of the electronic nose principle

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Summary. *As regards environment, the information to be provided to the decision maker or to the manager must be clear, accurate, unambiguous, and ideally it should be the result of the aggregation of a great number of data or parameters. The paper presents the concept of some "integrated index", already used to assess the quality of the environment. It stresses on the application of the "electronic nose" principle to the continuous monitoring of odour in the environment. The instrument response is a "pattern", similar to an integrated index, directly related to the annoyance, as felt by neighbouring people. It gives thus an information which can be handled by a manager, and which is more rich than individual pollutant concentration values.*

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Subject category: 8 (sensor systems)

Introduction

The environmental monitoring doesn't require specific techniques with respect to other measurements in medical laboratories, in industry or in the food sector :

- same sensor types
- same techniques of data storage or transmission
- generally, same kind of data processing tools and signal processing methods.

A first difference is that the environmental monitoring results from a multidisciplinary approach. One must generally use several techniques at the same time, requiring knowledge from different disciplines.

A second typical feature of the environmental monitoring is that it is essentially conducted in the field. That implies some constraints :

- the climate influences both the phenomena under investigation and the sensors or the instrumentation used;
- the electrical supply network is non available, which implies to use instruments with low power consumption or to carry very heavy batteries in the field;
- more generally, environmental monitoring entails the use of portable instruments, which must be able to provide the useful information directly to the final user in the field;
- a frequent difficulty is the deterioration of the material by the animals or by the vandalism.

A third and probably more important feature which characterises the environmental monitoring is the type of information which must be supplied to the final user, who is for example the person in charge of a municipality, or the security and environment officer in a company, or the manager of a waste water treatment plant, … That information must be clear, accurate and non ambiguous : generally it should be the result of the aggregation of many variables.

Indeed, for example, the concept of water quality is the integration of many parameters, such as the pH, the dissolved oxygen, the water conductivity, and so on, and it is the same for the air quality or even for the concept of "odorous annoyance".

To be relevant, the indicator should express the effect, or the pressure on the environment as a whole, in a single score.

Environmental indexes

Some such aggregate index are already found in the literature. For example, the "Organic Pollution Index" characterises the global organic quality of a river. It has different forms. One of them is the combination of 4 parameters [1] : the Biochemical Oxygen Demand and the concentration of Ammonium, Nitrites and Orthophosphates. According to its value, each parameter receive a class number, from 1 to 5, and the average value of those class numbers is the Organic Pollution Index : 1 means the most acute pollution and 5 means zero pollution. Unfortunately, the measurement of those parameters are only carried out in the lab, and some of them, like BOD, need 5 days to be evaluated. So, the global indicator can't be provided to the final user in real time, and sometimes it is too late to make a decision. Thus, such water quality index isn't really relevant for decision making purpose.

More conveniently, the index should be calculated from the values of classical variables, easy to measure.

A typical example is the thermal comfort, which is often measured to evaluate the indoor pollution.

The PMV (Predicted Mean Score) provides the objective thermal feeling of individuals who live in the environment being tested. In fact, it is based on the values of "environmental" parameters, such as temperature, humidity, air speed, radiant temperature as well as on values of the "personal" parameters : activity carried out by the occupants and their type of clothing. The more the result of the PMV index deviates from "zero" the more the occupants will have an uncomfortable thermal feeling.

Some commercial instruments exist to measure the PMV index in real time, we have also developed such instrumentation at FUL.

We have also applied a similar principle to the fog forecasting in the neighbourhood of the Meuse river, in Belgium. Eight climatic parameters were measured, those were rather usual parameters (temperature, humidity, wind speed), and the useful output variable was the probability of fog appearance 3 hours after the measurement.

Another application was the development of a ceramic sensor to measure the soil quality, which combines in the same body the measurement of three parameters : the water content of the soil, its salinity and its temperature. Again, the output useful variable, which combine those three parameters allows to assess a "global" soil quality factor.

Electronic nose principle

Some arguments justifie the development of an instrument able to monitor the odours generated in the environment.

- The odour, with the noise, appear among the most significant subjects of complaints in the public. However, until now, the odour is generally measured by the human nose and its interpretation is very subjective. So, it is justified to focus a research on the development of objective methods to measure the odours in the environment.
- Secondly, even if the odour cannot directly be regarded as a toxicity, it is always an alarm, precursor of pollution with more dramatic consequences. So, a continuous monitoring of the odour would allow to anticipate the rising of the toxic level.
- Thirdly, the odour is a reliable indicator of good or bad working of a process. In this case, an electronic nose could be used to monitor the odour, for example, with the aim of controlling some odour abatement system.
- And, finally, measuring an odour implies detecting gaseous compounds at very low concentration, sometimes well below the ppm level. Thus, the electronic nose principle can be generalised to the detection of other gaseous mixtures at low concentration, even if they aren't odorous. For example, we apply also the same principle to the monitoring of air pollution inside the buildings.

The electronic nose principle seems well suited to such objectives.

It is made of an array of non specific gas sensors. If all the sensor responses are put together, they form a pattern which is typical of the odour presented to the array, like a signature.

The electronic nose principle is characterised by a learning phase, during which a great number of gaseous mixtures are presented to the sensor array. Thanks to a pattern recognition engine, a library of typical patterns for the sensor signals is constituted, each of them corresponding to a specific environmental odour.

For our work, we have chosen so far to use tin oxide sensors, based on the change of electrical conductivity when a gas compound interacts with their surface. Their use is thus very simple. Their major drawback, for applications in the field, is that the sensing element must be maintained at an elevated temperature, typically above 300°C, in order to improve the kinetics of adsorption. And so, the sensor array consumes about 1 Ampere.

But, generally, tin oxide sensors are quite robust, their response is rather reproducible, and they are commercially available (the choice is to work with sensors from the Japenese company Figaro).

We have chosen to work with home-made instrument because commercial electronic noses are not suitable for environmental applications in the field : they are rather expensive and, generally, they are intended for laboratory applications for which the odour is sampled from the head space above a liquid or a solid.

In the field, the air is directly sampled in the gaseous ambience, with its humidity, and its temperature.

Also, the classical way of using the sensors is to work by cycling between a reference air, free from any contaminant, and the odorous sample. The response of the sensor is generally the difference between the signal, after equilibrium in the odorous ambience, and the base line generated in pure air.

But the use of a pure air cylinder is not convenient for field applications : that is heavy and cumbersome. Alternatively, filtering the ambient air through charcoal or molecular sieves needs very restricting maintenance conditions.

We decided not to work with perfectly pure air, but with the ambient air, coarsely filtered and not dried. The goal is not to create a base line for the measurement reference, but simply to purge the sensor vessel, so as to regenerate the sensors.

Obviously, the recorded signal is less pretty than the one obtained with a commercial electronic nose, under the rigorous operating conditions of the laboratory, and with perfectly controlled and stable head space.

Thus, the continuous monitoring of environmental odours in the field looks like a challenge with respect to laboratory utilisation. But the actual results in that area are promising and the potential of applications is enormous.

To become a reality, the use of electronic nose in the environment has first to overcome some difficulties, such as :

- the understanding and the control of the influence of ambient parameters (temperature, humidity),
- the improvement of sensors sensitivity and noise reduction in order to be able to detect the very low concentration levels of odorous compounds in the atmosphere,
- the identification of main environmental odours by typical signatures, in various operating conditions and non-constant odour intensity.

The used pattern recognition technique is either an unsupervised procedure, such as Principal Component Analysis, or a supervised procedure, like artificial neural network with backpropagation algorithm, or Discriminant Function Analysis (DA).

The unsupervised procedures are free to respond to input data and to build up a "model" which is able to cluster the observations into some groups showing similar behaviour with regard to the observed variables. We shall consider them as good evaluation tools for a performance evaluation of the system during the development phase.

Again, when other electronic nose users show very nice ellipses around well separated groups in the plane of the principal components, more scattered clouds are observed for environmental applications : the dispersion of the data is mainly due to the wide range of operating conditions.

But, anyway, the electronic nose principle, and the classical pattern recognition procedures are applicable in the field, because, if the results are worse, the requirements are less restricting. For example, in food analysis, one have to class very accurately three or four coffee beans, or edible oils, in order to grant a quality label to the product. The monitoring of a given odour in the environment aims just at detecting the rise of the odour in the background, in order to supply a warning signal when the odour increases beyond a given threshold : that is only an "on-off" signal, and not an accurate measurement.

Unlike unsupervised methods, supervised procedures have a teacher providing target groups, besides the input stimuli. For such procedure the input signals are put in relationship with odour sources, and the membership in a specific group is known.

After the learning phase, a model is calibrated, and a new item is then diagnosed by determining how typical its individual pattern of variables is of a given group.

We always use supervised techniques, when, at the end of the development level, we have to build such model, in order to recognise the odour in real time.

Some results

The purpose of the first experiments at FUL in Arlon was essentially to study the influence of the water content of the sample on the sensor signals and on the pattern recognition. Indeed, the odorous mixture generated by any industrial source may exhibit a water content ranging from zero to about saturation. Consequently, the semiconductor resistance variation is modified or even reversed when humidity changes. We used an array of 12 "Figaro" sensors in a 6 litres Perpex cubic chamber. Those experiments were carried out in the lab, in dynamic conditions.

We have first tested the recognition of synthetic odours, made of chemical compounds, typical of environemental odours : that is alcohols, esters, amines, aldehydes, ketones and sulfides.

Each sample is prepared in a Tedlar bag, but under uncontrolled external conditions, and thus under various humidity levels. We have tested here a neural network with 18 log-sigmoïd neurons and a backpropagation algorithm. For the network learning step, we used a training set of signals generated by the odour at any humidity level. In such conditions, the network is able to recognise 6 new samples. New samples means : 6 validation samples, not used for the calibration procedure.

In a second step, a similar operation is performed, based on the same samples, but this time, the training is performed only with samples at low water content. Then, the model so calibrated is tested for the recognition of more humid samples, but it is no more able to recognise all the validation samples.

Thus, those experiments show that, as long as sampling and learning are carried out under many different humidity conditions, and not under particular ones for a given source, the classification remains relevant, and the humidity may be considered as a "neutral" variable.

A second set of experiments concerns several real environmental odour sources.

Again, the tests are carried out in the laboratory with 12 Figaro sensors. The malodours are sampled in Tedlar bags, near the sources, thus at the emission level.

Five odorous sources are tested, they cover a range of typical environmental odours.

Those are :

– a rendering plant, in the vicinity of the oven,
– a paint shop in a coachbuilding, the sample is collected either during or after the primer spray painting work of a car door, inside the workshop,
– a waste water treatment plant, near the fresh sludge aerobic treatment work,
– urban waste composting facilities, near the compost deposit area, which is under a shelter,
– and a printing house (in fact there were two different printing houses for that study).

A total of 59 samples are collected during a period of 7 months, between March and October, in various climate conditions, and sometimes at various operating conditions.

Figure 1 shows the results of the discriminant functions analysis. The figure displays the results in the plane of the first two discriminant functions calibrated from 49 out of the 59 samples (10 samples are kept for validation purpose). Differences between sources can be clearly observed. Only the "rendering" cluster is very close to the "compost" cluster, but it is normal : the two odours are very similar (both contain ammonia, aldehydes, alcohols and fatty acids).

Moreover, the 10 samples kept for validation purpose are well classified by the calibrated model (the big symbols).

Two additional samples are also tested, they are labelled as "bugs". They are collected in the waste water atmosphere, but during the sprinkling of chemicals masking the odour. We can see that these two "special" points are not recognised as belonging to the "waste water" group : that is an encouraging result.

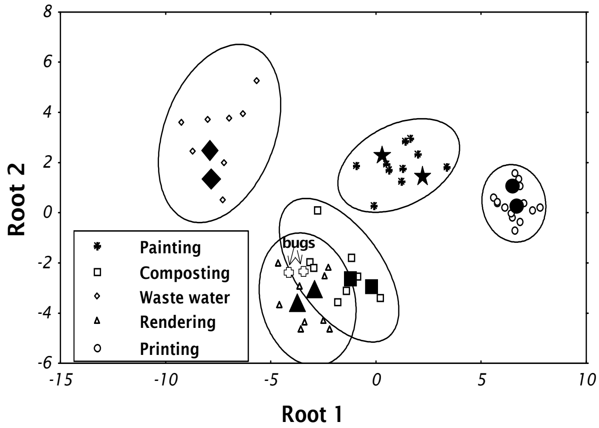

Fig. 1: Classification model calibrated by DA for 5 environmental sources : right validation of 10 unknown samples (two samples for each five odour : big symbols), no recognition of the two "masked" wastewater odours (labelled "bugs")

Another interesting point is that, for those classification procedures, the "signal" used for each sensor is $(1-R_{norm})$ where R_{norm} is the normalised resistance, or the resistance of the sensor, divided by the quadratic mean of all sensor resitances. The net effect of that normalisation is to reduce the dependency of the odour concentration from the array response, and also to reduce a little bit the effect of the sensor drift. That is a very classical way of signal pre-processing. But, what is less classical is the fact that the best classification is always obtained with the raw resistance, and not with the difference between the signal and the base line (i.e. : the response to the reference air). Obviously, such result is in opposition to the general advice found in the literature. The reference to the base line is known as always improving the signal, as it reduces the influence of drift or poisoning. However, in this case, as the "reference" is not a very pure dry air, the small variations of the concentration of trace elements in the reference air influence the classification. It is an important result : that means that avoiding the reference to the base line should lead to satisfactory classification results, and that the use of gas cylinder or high quality filters could perhaps be avoided. Anyway, the sensors need obviouosly some "reference" air to regenerate, but not really "pure" air, and not necessary before each measurement.

However, the problem of sensor drift is not solved yet : for long term studies (more than 6 … 8 months), a periodical calibration of the sensor array with suitable reference gas should be necessary.

Figure 1 shown the classification of the 5 odorous sources by the discriminant analysis. But for practical purpose, it is not needed to distinguish between 5 sources. A more plausible situation should be to detect the rising of an odour in the background. The figure 2 shows the result of a discriminant analysis carried out with the 59 observations in the surrounding of the sources and with an equivalent number of observations made in odourless air.

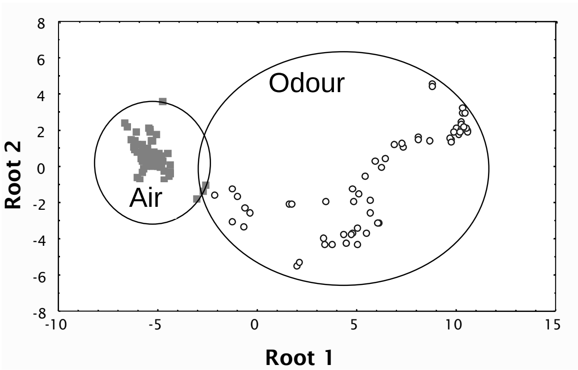

Fig. 2 : Discriminant model calibrated by DA for 59 observations in the surrounding of odorous sources and 59 observations in odourless air.

Such model is able to detect the presence of any odour with respect to odourless air : that seems an obvious result, but it is sometimes sufficient for many applications.

Moreover, such result can already be obtained with a quite simple device. In order to test the feasibility of a field electronic nose, we have also used a "mobile detector" simply made of an electronic board on which 8 tin oxide sensors were soldered. The sensors are heated by a battery-powered power supply. They are simply in static contact with the ambient odorous air. The measurements are made with a portable data logger in the field after stabilisation, and several times during the test period.

Even with that very simple device, it is possible to classify the 5 sources with a relatively good accuracy.

But, in this case, the main problem is the influence of the air movement around the sensor, which modifies the conditions of heat convection, and thus the temperature of the sensor.

So, a portable instrument has been developed, made of 6 TGS sensors in an aluminium vessel of about 100 cm^3. A constant gas flow rate of 150 ml/min is provided by a small pump, and the system operates by a series of cycles, alternating 5 minutes of "air" (sampled in a Tedlar bag from ambient air, far from the source), and 5 minutes of odorous gas transferred directly into the sensor chamber. The whole system is powered by a 12 Volts battery.

With such mobile detector, very promising results were obtrained in the spirit of a continuous monitoring. One of them will be illustrated here. During a first learning phase, we have presented to the detector the 5 odorous sources already mentioned, or just odourless air. From those measurements, we calibrated a model based on the discriminant analysis. Later on, we used the so calibrated classification functions to recognise the odour when the detector was moved around the print shop. Figure 3 shows the result.

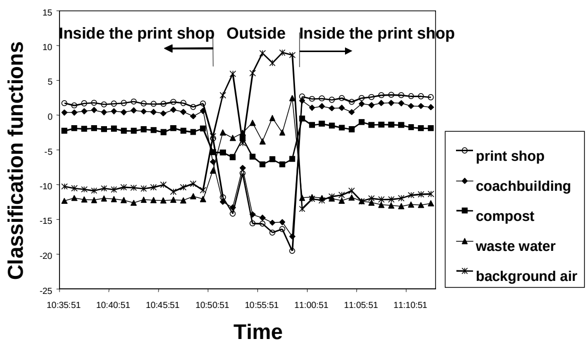

Fig. 3 : Evolution of the calibrated classification functions when the detector is moved away from the source in the print shop.

The scaling of the horizontal time axis is unessential : it shows only that the mobile detector is continuously moved away from the source.

The figure shows that the classification function corresponding to the printing shop has effectively the highest value when the detector is in the printing shop, but that it suddenly drops when the detector is moved outside the shop, and increases again when the operator moves again inside. As expected, the classification function characterising the odourless air (here named "background") is higher when the electronic nose is outside.

Such finding is promising : it proves that it could be possible to detect the rise or the vanishing of a particular odour in the background. Thus, to monitor a specific odour, even when there are some other influences, like the passing of a truck, or some other disturbance.

Finaly, two other interesting applications in the field are presented hereafter.

The first one concerns the assessment of the odour generated by a landfill of urban waste. Three kinds of odours are perceived by the neighbouring population : either the one of the fresh refuse (esters, sulphur organic compounds, solvents, …), or the one of the biogas generated by the decomposition of the organic matter under anaerobic conditions (trace elements, such as H_2S, NH_3 and some VOC's in a mixture essentially composed of odourless compounds : methane and carbon dioxide), or the one of clinker, which is the residue of waste incineration and which is characterised by a pungent odour.

During the sampling time, the technician tried to identify the odour with his nose. He recognised the biogas odour 9 times, the clinker odour 6 times and the fresh waste odour 9 times (these observations correspond to the same number of observations with the electronic nose). The 24 groups of responses to the 6 sensors constitute the input data set of a Discriminant Analysis, together with 27 additional measurements made on pure air. The feeling of the technician for each observation is used to specify the target group.

Figure 4 shows that the first root of the calibrated model separates well the odorous samples from the air ones. The second root distinguishes rather well the different odour sources. Such results are preliminary : future works will focus on the same application directly in the field, with a mobile detector aiming at detecting and identifying in real time the various odours generated in the landfill.

Indeed, the final goal, corresponding to the wish of the landfill manager, should be to use continuously the calibrated model in the field in order to control the atomisation of the bad odour neutralisation product.

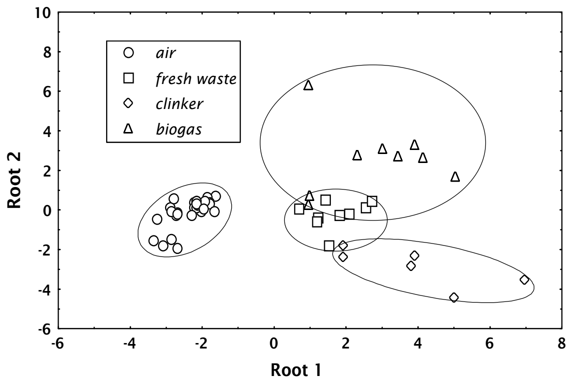

Fig. 4 : Discriminant analysis on the odour generated by a landfill of urban waste.

The last application concerns the monitoring of indoor air pollution.

The department "Environmental Monitoring" at FUL collaborates with a service of investigation of indoor pollution. They measure some pollutants in buildings, and for some of them, the only analysis method is the GC-MS at the laboratory level from samples of the ambient air. However, the sample is not always representative of the typical ambience : may be a window was just opened before the sampling, or somebody entered in the room with a perfume, or there are cooking vapours, …

So, they ask us to develop an instrument to monitor continuously the ambience in a room, so as to point out a warning signal when a given pollutant rises above a threshold. That signal may serve to switch on the sampling device.

For that application, we were specially aware of the ambience of solvents in the buildings (benzene, toluene, xylene, and other ones), and we have used a multisensor from the Swiss manufacturer Microsens. Those sensors are based on thin-film, metal oxide technology. They are packaged in standard TO8 metal can (12 pins) and each package contains either 4 or 6 sensors.

We have tested so far the response of the sensor to very low concentration of BTX (lower than 1 ppm), and now, we are conducting field test. Each time, the signals provided by the multisensor are recorded, and in the same time, a sample of the ambient air is taken and analysed by GCMS. The aim is to find a relationship between the pattern formed by the sensor signals and the concentrations of solvents. That research is now going on, and it seems to be a very promising application of the electronic nose.

Conclusion

The various tests conducted in the field with home-made electronic noses, with very simple configurations, lead to very promising results. They allow to explicit the specifications of a portable instrument, able to predict an unknown odour in the environment, and to monitor it continuously, on the basis of the classification model calibrated during the learning phase.

The monitoring of environmental odour is really a challenge, but for environmental use, there is no need for very accurate response : a rough detection is sufficient for most applications.

Consequently, there is no need for operating conditions as restricting as laboratory electronic nose ones.

Such conclusions are encouraging, but further works are still needed before reaching the final goal and to point out the limits of the method.

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