A MODEL BASED ON ARTIFICIAL NEURAL NETWORK FOR RISK ASSESSMENT TO POLYCYCLIC AROMATIC HYDROCARBONS IN WORKPLACE

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ABSTRACT

Polycyclic aromatic hydrocarbons (PAHs) are formed during incomplete combustion in different production processes; exposure to PAH-containing substances increases the risk of cancer in humans.
The environmental monitoring used to assess human exposure to airborne PAHs during work, generally involves the employment of diagnostic methods derived from analytical chemistry, characterised by an elevated cost and the use of a "trial and error" approach.

The aim of this study is to develop a decision support tool that, through the characteristic parameters of a workplace and using an artificial neural network, simulates the concentration of different species of pollutants (PAHs groups) statistically present in the environment. In this way it is possible to perform a preliminary risk assessment that, besides allowing an immediate perception of the level of risk to which workers are exposed, can undertake environmental monitoring analysis on the detection of a limited number of pollutant species, in order to reduce costs and increase the sustainability of the production system.

Keywords: model of prediction, risk assessment, environmental monitoring, decision support system

1. INTRODUCTION

There is a growing interest, both scientific and industrial, in the environmental management of production systems. Great attention is paid to activities that increase the sustainability of production systems.
In this context, there is significant interest in workplaces in which air pollutants that are highly dangerous to the health of workers are emitted by production processes. Among the various types of pollutant, Polycyclic Aromatic Hydrocarbons (PAHs) are amongst the most harmful chemical compounds to human health. The carcinogenic effect of PAHs has been deeply investigated in the past, and is nowadays well known.

Considering the large variety of production processes that result in the generation and dispersion of pollutants, there is a wide range of monitoring equipment based on technologies derived from analytical chemistry. So it is often very difficult to perform preliminary risk assessments of a specific workplace. In fact, in many cases the analytical methods used for environmental monitoring are expensive and use a "trial and error" approach.

Forecasting the concentrations of air pollutants represents a difficult task due to the complexity of the physical and chemical processes involved. Several approaches have been used, branching into two main streams: deterministic approaches, which involve numerically solving a set of differential equations, and empirical approaches, where different functions are used in order to approximate the concentrations of the pollutants depending on the external conditions (Hrust et al., 2009).
The first type of approach does not require a large quantity of measured data, but it demands sound knowledge of the pollution sources, the temporal dynamics of the emission quantity, the chemical composition of the exhaust gasses and physical processes in the atmospheric boundary layer. This crucial knowledge is often limited and also requires computational resources. Thus approximations and simplifications are often employed in the modelling process. On the other hand, applications of deterministic models are limited to a lesser extent with regard to the selection of the domain. A recent example of such an approach is the work of (Finardi et al. 2008).
In contrast, the second type of approach usually requires a large quantity of measured data collected under a large variety of atmospheric conditions. By applying regression and machine learning techniques, a number of functions can be used to fit the pollution data in terms of selected predictors. One drawback of this technique is that the model is usually confined to the area and conditions present during the collection of the measurements (Kukkonen et al., 2003). Nevertheless, this approach is generally more suitable for the description of complex site-specific relations between concentrations of air pollutants and potential predictors, and consequently it often results in a greater accuracy, when compared with deterministic models (Gardner and Dorling, 1999).

Neural network empirical approaches have been frequently used in recent atmospheric and air quality modelling studies. (Božnar et al. 1993) were the first to

describe the neural network modelling of hourly concentrations of sulphur dioxide. (Gardner and Dorling 1998) gave a very informative review of the applications of artificial neural networks in science in general and, particularly, in atmospheric sciences. They emphasised the usefulness of neural networks (NN) when dealing with non-linear systems, especially when theoretical models of the system cannot be constructed. (Perez et al. 2000) developed a neural network model to predict PM_{10} hourly concentrations by fitting a function of 24-hourly average concentrations from the previous day. They found errors ranging between 30% and 60%. In order to decrease the errors, they considered noise reduction in the data, rearrangement, an increase in the learning dataset, and the inclusion of meteorological variables as input variables. They concluded that noise reduction prior to modelling is essential. A possible improvement can be achieved by explicitly taking into account the relevant meteorological variables.

Karppinen et al. published two papers addressing the development of a modelling system for predicting NOx and NO_2 concentrations in an urban environment in Helsinki. The first paper (Karppinen et al., 2000a) was related to the model development and its application in air quality prediction, as well as traffic planning. The system includes the following models: the estimation of traffic volumes and travel speeds, the computation of emissions from vehicular sources, a model for stationary source emissions, a meteorological pre-processing model and dispersion models for stationary and mobile sources. Alternatively the second paper (Karppinen et al., 2000b) presented a comparison between the predicted and measured concentrations. According to the authors, the modelling system was fairly successful in predicting NOx concentrations and was successful in predicting NO2 concentrations. They also argued that none of the methods are able to forecast the peak values, due to the under-representation of these cases within the overall dataset.
(Perez and Reyes 2006) developed a multi-layer perceptron (MLP) model to forecast the daily maxima of PM_{10} concentrations one day in advance. The same model was applied to five measuring stations in the city of Santiago, Chile. They compared values forecasted with MLP, linear and persistence models using the same input variables. They concluded that the MLP model performed well and that the relatively small differences between the linear and MLP models emphasised the importance of selecting the correct input variables.

In the scientific literature there are only a few cases related to the use of predictive models in confined spaces, such as apartments, residential buildings, workplaces, etc.
The aim of this work consists in defining a support decision tool for preliminary risk assessment in workplaces, where is possible to identify a narrow set of environmental variables, and there is a strong correlation between concentrations of pollutant in the air and the typology of the manufacturing process. For this scope an Artificial Neural Network (ANN) has been realised that is able to provide a forecast about the presence and concentration of the main pollutants released by a particular production process, in connection to different distances from the source of emission.

This paper is structured as follows: section two deals with a taxonomic clustering of the industrial processes that provide the production of similar outputs, also characterised in many cases by the same emission levels. In section three a model is proposed that, depending on the input parameters such as the total PAHs, distance and categories of workplace, can provide prediction data for the relative concentrations of different pollutants. Finally, the model results are examined in order to evaluate the reliability of the forecast generated by the model.

2. WORKPLACE: TAXONOMIC EVALUATION OF SOURCE PAHS

Polycyclic aromatic hydrocarbons (PAHs) are formed during incomplete combustion. They occur in the environment as complex mixtures of many components with widely varying toxic potencies. Several compounds of this group have been classified by the International Agency for Research on Cancer (IARC) as probable (2A) or possible (2B) human carcinogens (Boström, 2002). Due to its high carcinogenic potency and its presence in the environment, benzo(a)pyrene (Bap) is often used as an indicator of human PAHs exposure (Han, 2011). Road paving, sintering plants, and rubber production are only a few of the principal workplaces where there are industrial processes that emit high concentrations of PAHs into the environment.

The first phase of this study consists in the classification of workplaces into four categories. Each category includes industrial processes that provide for the production of an output that is similar or complementary to the same supply chain.

The categories identified are as follows:

- *Energy activities* that include all those processes related to the production of energy, including: combustion plants, oil refineries and gas, coke ovens, gasification plants and liquefaction of coal;
- *Production and processing of metals* which include: production plants of cast iron or steel, plants for the processing of metals, foundries, sinter plants and surface metal treatment plants;
- *Cooking activities* that include: industrial kitchens and restaurants;

- *Insulation activities* that include: treatments of roofing bitumen for buildings and road paving.

For all categories a series of case studies from the literature have been analysed, and for each the measures of concentration of particulate-bound PAHs emitted into the environment have been considered.

To assess the health-risks associated with PAHs exposures, it is important to know the total carcinogenic potency arising from the exposures of various PAHs compounds. In principle, the carcinogenic potency of a a given PAHs compound can be assessed according to its benzo[a]pyrene equivalent concentration (BaP_{eq}). Calculating the BaP_{eq} concentration for a given PAHs compound requires the use of its toxic equivalent factor (TEF; using benzo[a]pyrene as a reference compound) to adjust its original concentration. Among the 16 PAHs species identified as priority pollutants, we consider seven PAHs species absorbed on particulate matter, according to the carcinogenic potency factor developed by the US Environmental Protection Agency (Table 1), including: Benzo[a]anthracene (BaA), Chrysene (CHR), Benzo[b]fluoranthene (BbF), Benzo[k]fluoranthene (BkF), Benzo[a]pyrene (BaP), Indeno[1,2,3-c,d]pyrene (IND), Dibenz[a,h]anthracene (DBA).

For each case study the corresponding BaP equivalent concentration (BaP_{eq}) has been calculated using the TEF approach recommended by the US EPA.

Table 1: Benzo(a)pyrene Toxic Equivalent Factor (US EPA, 1986)

Compound	Carcinogenic potency
Benzo[a]anthracene (BaA)	0.1
Chrysene (CHR)	0.001
Benzo[b]fluoranthene (BbF)	0.1
Benzo[k]fluoranthene (BkF)	0.01
Benzo[a]pyrene (BaP)	1
Dibenz[a,h]anthracene (DBA)	1
Indeno[1,2,3-c,d]pyrene (IND)	0.1
Other 9 PAHs species	0

It has been observed that categories related to activities relating to insulation and energy are characterised by the highest values of Bap_{eq} (about 800 ng/m3). In fact bitumen is a complex hydrocarbon material containing components in many chemical forms, the majority of which are of high molecular weight (Posniak 2005). In experimental studies polycyclic aromatics with 3 to 7 fused rings with molecular weights in the range 200 to 450 have been shown to be biologically active carcinogens, in particular Benzo(a)pyrene and Dibenz[a,h]anthracene (present in a high percentage) are considered to be powerful carcinogens (Agency for Toxic Substances and Disease Registry, 2009). Moreover fumes, created when asphalt is heated, contain very small, solid, airborne particles that are easily inhaled by workers. Fumes may also contain hydrogen sulphide vapours, which are very toxic, as well as vapours generated by the solvents used to "cut" the asphalt (Burstyn 2000).

In the combustion processes, for the production of energy, the main cause of emissions to the atmosphere of particulate-bound PAHs is coal. Coal in fact contains large quantities of organic and inorganic matter. When coal burns, chemical and physical changes take place, and many toxic compounds are formed and emitted; PAHs are among those compounds. The emissions, in this case, are limited by filtering systems present along the lines of the process (Boström 2002).

The category for the production and processing of metals is characterised by the intermediate values of Bap_{eq} (of the order of 50 ng/m3). The mechanisms associated with the generation of PAHs in the high-temperature combustion process of the smelters works, followed three major pathways, including pyro-synthesis, direct emission of unburned fuel, and thermal destruction of fuel components (Tsai, 2001). The PAHs formed and released by pyrolysis in a limited oxygen supply can appear free in gaseous form and are adsorbed onto dust particles (Tsai, 2000). For the iron and steel industries, PAHs are released from coke manufacturing, sintering, iron making, casting, moulding, cooling, and steel making processes (Lin, 2008). The average emission levels are lower than the values of the preceding categories, because this process requires a lower percentage of organic matter, compared to the activities of insulation and energy.

The category that includes cooking activities is the least dangerous when compared to the other categories; in fact emissions of PAHs in this workplace are in the order of 10 ng/m3. In industrial kitchens or in the restaurants, the emission of PAHs is related to frying at high temperatures. The levels of the (PAHs) pollutants, from the process of cooking, are strongly related to cooking style, lipid content of the food, and the quantities of food cooked. Another variable that significantly affects the level of PAHs emission is the type of cooker, in fact frying on a gas stove caused significantly higher amounts of ultrafine particles compared with frying on an electric stove (Li, 2003). If on the one hand this category includes workplaces with smaller dimensions compared to the workplaces of the other categories, on the other hand the rate of natural ventilation is higher than in non-residential buildings.

3. MODEL OF PREDICTION FOR PAHS EMISSIONS

The second step in this work consists of defining the input and output parameters of the model.

The model includes indoor workplaces or those activities that imply a direct contact among the sources of emission and the worker (e.g. workers employed in road paving, manufacturing and laying of bituminous mixtures, etc.). The input variables of the model are as follows:

Total PAHs: it is a numeric variable given by the sum of the concentration (only the particulate matter phase)

of all priority compounds, belonging to PAHs groups, that are the most dangerous to human health;

Distance: it is a qualitative variable that as a function of the distance between the sources of PAHs emission and the position of devices for environmental monitoring can assume three different parameters:

1. First Line: distance between the source of PAHs emission and the device for environmental monitoring is less than 2 metres;
2. Second Line: the distance between the source of PAHs emission and the device for environmental monitoring is between 2 and 10 metres;
3. Third Line: the distance between the source of PAHs emission and the device for environmental monitoring is over 10 metres.

Categories of workplace: this is a qualitative variable that consider the type of manufacturing process or activity from which the PAHs emissions are generated. This variable can assume four types corresponding to: energy activities, production and processing of metals, cooking activities or insulation activities, as already described in the previous paragraph.

The core of the prediction model consists of a system based on an Artificial Neural Network (ANN). This learning technique, that mimics the biological learning process occurring in the brain, has been used. Neural networks present a robust way to predict real-value concentrations after learning from a supplied sample set. Such networks connect a number of individual elements, each of which take a set of inputs and produce a single real number. The learning algorithm determines the numeric weights to be applied between each of these neurons to obtain the desired output. One main advantage of this technique is that it can produce good results, even when supplied with noisy and incomplete data (Aquilina, 2010).
The network architecture provides: three nodes in the input; three hidden layers consisting, respectively, of 2-12-21 neurons and seven nodes of output (Fig. 3). Each input signal is weighted, it is multiplied by the weighted value of the corresponding input line (by an analogy to the synaptic strength of the connections of the biologic neurons). The artificial neuron will combine these weighted inputs by determining their sum, and with reference to a threshold value and an activation function it will determine its output. In this case a *Gaussian distribution* is used for assigning the weights to each variable.

The ANN works with a data set identified by a sample, i.e. a subset of the population representing the phenomenon studied. To be more precise, given the ANN three types of subset of the available sample can create the forecasting model: the training set, the test set, and the validation set:

- *training set*, the group of data constituted by a sample of 60% (percent of total data) that train the network, i.e. by which the network adjusts its parameters (thresholds and weights), according to the gradient descent for the error function algorithm, in order to achieve the best fit of the non-linear function representing the phenomenon;
- *testing set,* the group of data constituted by a sample of 20% (percent of total data), given to the network still in the learning phase, by which the error evaluation is verified in order to effectively update the best thresholds and weights;
- *validation set*, the group of data constituted by a sample of 20% (percent of total data) used to evaluate the ANN generalisation, i.e. to evaluate whether the model has effectively approximated the general function representative of the phenomenon.

The network has been trained using the back-propagation routine. This typology of algorithm is used to train a network for a desired output. This method minimises the squares of the residuals (differences between desired outputs and network outputs) by modifying the network weights. It approximates the desired output using the gradient descent technique.

The validation data set has been used to monitor the alteration in the training error during the learning progress (Fig. 1) of the neural network.

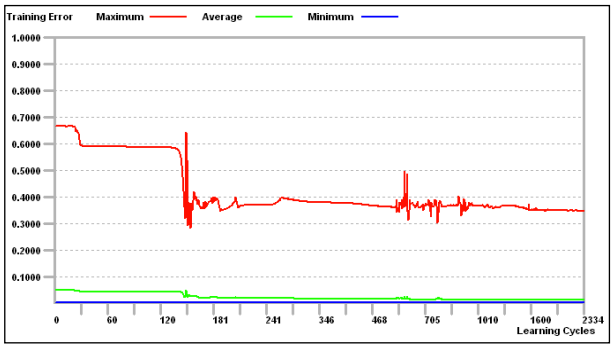

Figure 1: Alteration of the training error

In order to minimise the overtraining problem, the training phase is stopped when the mean square error (MSE) assumes values lower than 0.01.

To evaluate the accuracy of this ANN, the correlation coefficient between the measured data (training set) and the data predicted by the trained neural network (Fig. 2) has been calculated.

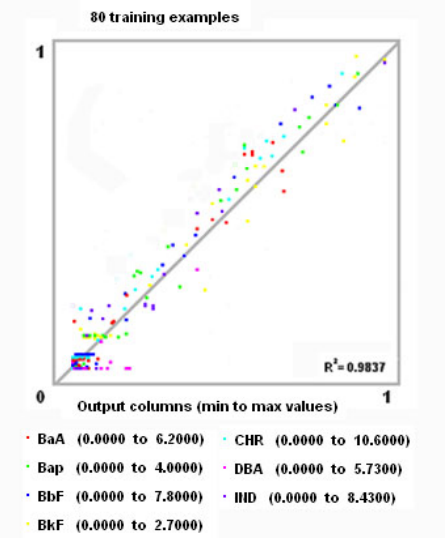

Figure 2: Forecast of the ANN (on training set)

According to US EPA guidelines, among the 16 PAHs species identified as priority pollutants, the model includes only seven PAHs species absorbed on particulate matter, that are considered powerful carcinogens (Tab. 1). The concentration, measured in the environment, of this species representing the output variables of the model (Fig. 3):

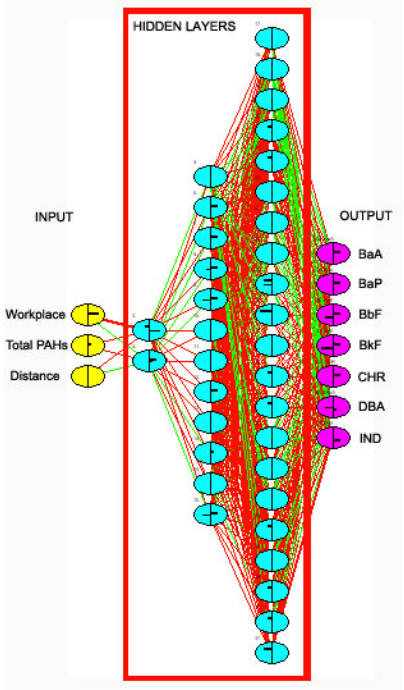

Figure 3: Scheme of the model

Based on this model it is possible to determine the level of concentration of individual PAHs pollutants, considered the most dangerous, in the environment. According to TEF defined by the US EPA, for each species of the PAHs group (Tab. 1) it is possible to calculate the concentration of B(a)p_{eq} in a specific workplace.

In this way, we have a direct perception about for the health risk assessment of workers exposed to PAHs particulate matter in the air.

3.1. Analysis of results

The model is tested using a series of data: for each category two set of data were collected for each of the three input variables that identify the distances between the sources of PAHs emission and the position of the devices for environmental monitoring.
Therefore, for each category and for every PAHs species identified as priority pollutants, the values of the concentration measured by analytical methods (shown as "real" in the following graphs) and those obtained by the prediction model (shown as "forecast") have been compared.

For energy activities (Fig. 4) it is observed that the gap between the real and forecasted values increases for higher levels of concentration. In fact, a Mean Squared Error (MSE) of about 0.02 has been calculated for values of concentration lower than 1 ng/m3, on the other hand for values of concentration higher 1 ng/m3 a MSE of approximately 2 has been calculated.

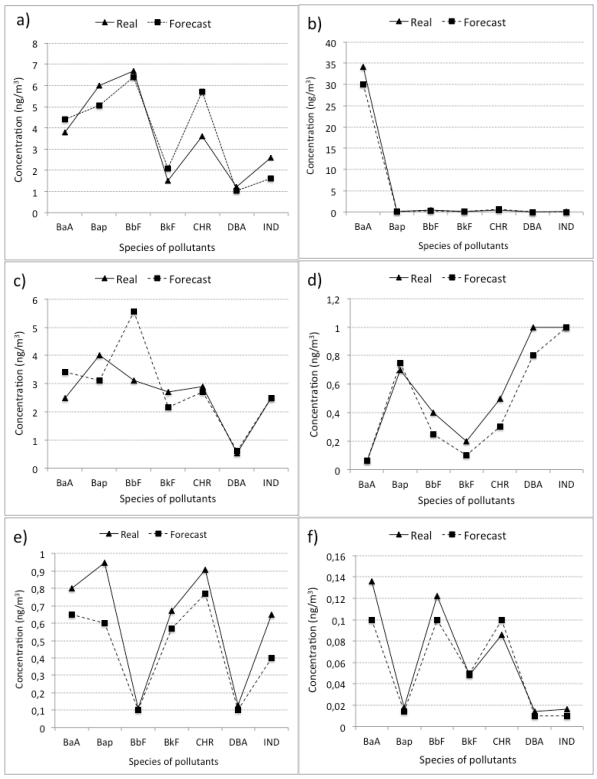

Figure 4: Comparison between real and forecasted values of concentration for single species of PAHs groups, in different scenarios of the "energy activities" category, in: first line (a-b); second line (c-d) and third line (e-f).

In order to evaluate the reliability of the forecast, the Mean Absolute Percentage Error (MAPE) has been calculated. For the first category of workplace the MAPE equals 24.65%.

The MAPE obtained for the category identified as *Production and processing of metals* amounts to 29.33%. In the following charts (Fig. 5) it is possible to assess the gap between value of the concentration measured by analytical methods (real), and the values generated by the prediction model (forecasts). Naturally these cases, like in the previous category of workplaces, are not included in the data set used for the training of the ANN.

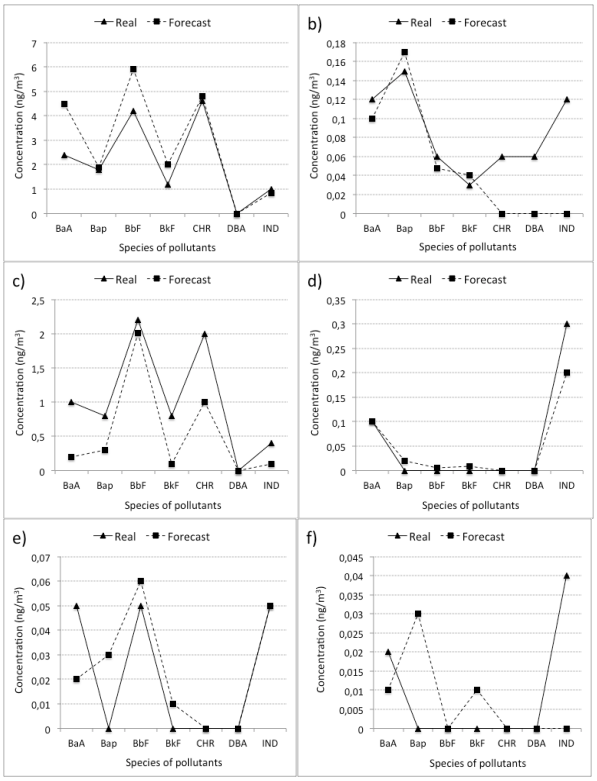

Figure 5: Comparison between real and forecasted values of concentrations for single species of PAHs groups, in different scenarios of the "production and processing of metals" category, in: first line (a-b); second line (c-d) and third line (e-f).

The evaluation of the forecasts for *cooking activities* (Fig. 6) shows that the prediction model reported a MAPE with a similar value as the other categories, in fact it calculated a MAPE equal to 23.03%.
For this category no measure of the concentration is available, with analytical methods, for a distance (between the source of PAHs emission and the device used for environmental monitoring) of over 10 metres.

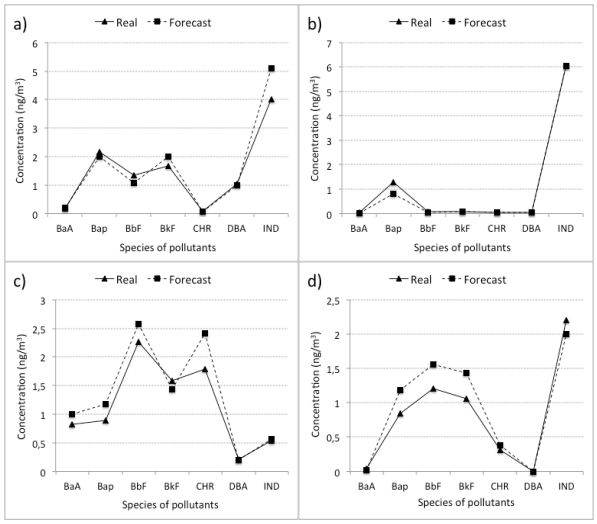

Figure 6: Comparison between real and forecasted values of concentrations for single species of PAHs groups, in different scenarios of the "cooking activities" category, in: first line (a-b) and second line (c-d).

The forecast generated by the model, for the last category, *Insulation activities* (Fig. 7), is more reliable compared to the predictions obtained for the other categories; in fact for this category of workplace a MAPE equal to 10.79% has been calculated. It is believed that this is due to the types of work process that fall in the category of insulation activities. In fact, in this category the activities are very similar to each other, all all characterised by the same temperatures, raw materials, combustion processes and boundary conditions. Indeed, this is not true for the activities belonging to the other categories whose work processes and boundary conditions may vary significantly from one working environment to another.

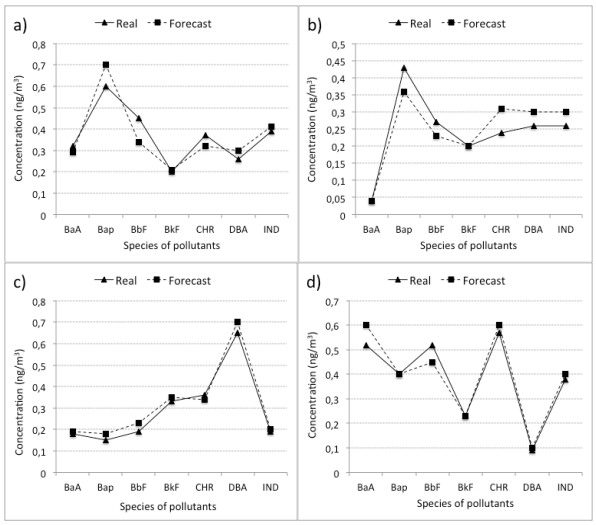

Figure 7: Comparison between real and forecasted values of concentrations for single species of PAHs groups, in different scenarios of the "insulation activities" category, in: first line (a-b) and second line (c-d).

Besides the evaluation of the MAPE for each category of workplace, the MAPE for all species of pollutants, independent of the type of industrial process (Tab. 2), has been calculated.

Table 2: Mean absolute percentage error for compounds considered most dangerous to human health

Compound	Carcinogenic potency	MAPE (%)
Benzo[a]anthracene	0.1	27.84
Chrysene	0.001	23.26
Benzo[b]fluoranthene	0.1	23.87
Benzo[k]fluoranthene	0.01	19.58
Benzo[a]pyrene	1	19.21
Dibenz[a,h]anthracene	1	14.66
Indeno[1,2,3-c,d]pyrene	0.1	25.22

It is possible to observe from the previous table that, according to carcinogenic potency determined by the US EPA (Tab. 1), the most dangerous compound to human health, among the PAHs components, are Benzo[a]pyrene and Dibenzo[a,h]anthracene (TEF equal to 1). The forecast of the model of these compounds is characterised by a MAPE of about 17% .

Calculating the $B(a)p_{eq}$for each category of workplace using the values of concentration, both real and forecasted (Fig. 8), it has been observed that a higher gap has been detected between "$B(a)p_{eq}$real" and "$B(a)p_{eq}$ forecast" for a distance, among the sources of PAHs emissions and devices for environmental monitoring, of over 2 metres (second and third line).

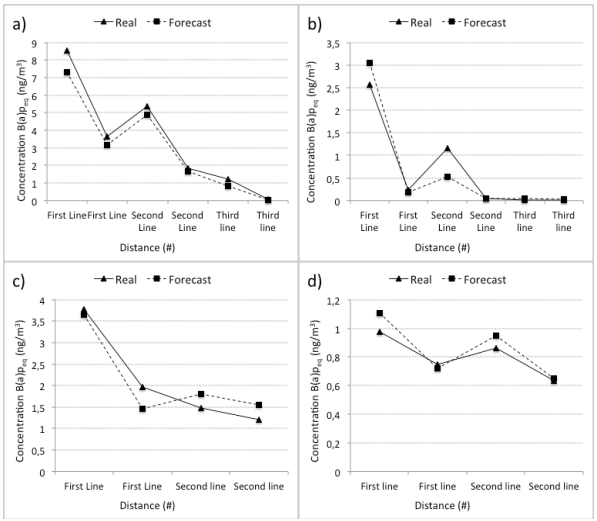

Figure 8: Comparison between real and forecasted values of concentrations of $B(a)p_{eq}$ for different distances, in cases of: Energy activities (a); Production and processing of metals (b), Cooking (c) and Insulation activities (d).

The evaluation of the MAPE, for different distances of each category, is summarised in the following table:

Table 3: Mean absolute percentage error for different distances and categories of workplace

Workplace	First Line (%)	Second Line (%)	Third Line (%)
Energy (a)	13.45	9.14	28.95
Prod. (b)	20.62	40.58	30.83
Cook. (c)	14.44	26.29	-
Insulat. (d)	8.54	6.04	-
Average	**14.26**	**20.51**	**29.89**

A lower MAPE has been calculated for "first line"; namely when the distance between the source of PAHs emission and the device for environmental monitoring is less than 2 metres. This is the most dangerous case for human health; because the level of the concentration of the pollutants to which the workers are exposed is the highest. The model, in this case, is able to ensure the best reliability of the forecast.
Significant MAPE values are due to a limited number of case studies from the scientific literature. However, the 'learning' capability of the ANN will provide more and more reliable results, provided that new training cases are available. To this end, simulated cases by specialised software (e.g. NIST [x]) could also represent a good opportunity to enrich the 'knowledge' of the ANN. Under this perspective the computer-based tool is found to be even more effective in predicting PAHs concentration values, thus avoiding or reducing time-consuming and expensive field investigations for the preliminary assessment of work environments as well as continuous monitoring.

CONCLUSIONS

The model is an efficient and economically sustainable tool: efficient because it can improve the performance with learning in time through the increase in the training data set. It is indispensable for a preliminary risk assessment and the environmental monitoring of a specific workplace.

It is economically sustainable because the model can orientate the decision making process toward the identification of a limited number of PAHs compounds (whose presence is statistically noted). In this way it is possible to reduce the costs of monitoring environmental performance with analytical techniques. In fact, if the prediction of the model does not indicate the presence of a single compound it is not necessary to measure its concentration with analytical techniques. In doing so the costs can be reduced by 20% for each compound not detected. Assuring, however, a high level of protection to the workers' health.

Moreover, performing environmental monitoring in parallel with the forecasting model and analytical methods, it is possible to reduce, in time, the sampling frequency of the analytical method. Reducing, once again, the costs of environmental monitoring and ensuring that the control of health in the workplace is efficiently and economically sustainable.

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