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Comparative analysis of maximum daily ozone levels in urban areas predicted by different statistical models

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ABSTRACT: Many large urban areas experience elevated concentrations of ground-level ozone pollution, which is reported to cause adverse effects on human health and the environment. The prediction of ground-level ozone is an important topic, which attracts attention from research communities and policy makers. This study investigates the potential of using the multi-layer perceptron (MLP) neural network technique to predict daily maximum ozone levels in the Bangkok urban area. The MLP was trained and validated using ambient air quality monitoring data and observed meteorological data for the high ozone months (January to April) in the area during a four year period, 2000–2003. The inputs to the MLP included the average concentration of air pollutants (nitrogen oxide, nitrogen dioxide, and non-methane hydrocarbon) and meteorological variables (wind speed and direction, relative humidity, temperature, and solar radiation) during the morning rush hours. The MLP network, which contained 8 input layer neurons, two hidden layers (10 hidden neurons for the first hidden layer, 14 hidden neurons for the second hidden layer) and 1 output layer neuron, was found to give satisfactory predictions for both the training and validated data sets. The performance of the MLP was better than the multivariable linear regression model developed based on the same dataset. For the validated dataset, the MLP predicted the daily maximum 1-h ozone concentration in the study area with a mean absolute error of 10.3 ppb, a root mean square error of 13.5 ppb, a coefficient of determination (R^2) of 0.85, and an index of agreement of 0.89.

KEYWORDS: ozone pollution, artificial neural network model, multivariate regression, Bangkok

INTRODUCTION

Ground-level ozone (O_3) has recently become a serious air pollution problem in many urban areas around the world. Numerous studies indicate that exposure to an elevated concentration of O_3 air pollution is a potential human health hazard[1–3]. O_3 also affects vegetation adversely. The negative effects may relate to visible foliar injury and to physiological impairment which consequently lead to a significant reduction in growth and yield of agricultural crops and forests[4–7]. Accurate prediction, with a long enough forecast time span, of O_3 air pollution in an area is important as it can produce necessary warning signals to help in reducing exposure and minimize adverse effects on humans and the environment.

As for any other air pollutants, O_3 can be predicted using statistical models or deterministic models. Deterministic models are based on a fundamental mathematical description of atmospheric physical and chemical processes[8–10]. These models, which numerically solve the complete set of time-dependent equations incorporating complex photochemical reaction mechanisms, have been used to study pollution of photochemical smog in urban areas, regional-scale dispersion of chemical species, long-range transport, and others. The deterministic models are highly sophisticated because they require a high level of human resources and powerful computers. Most importantly, these models require detailed and accurate emission data and meteorological inputs that are not commonly available in many regions. If these inputs are only available with large uncertainty then the results of deterministic models are questionable.

Statistical models are based on semi-empirical statistical relations among available data and measurements. They do not necessarily establish deterministic cause-effect relationships. They attempt to determine the underlying relationship between sets of input data (predictors) and targets (predictands). Many different statistical techniques have been proposed to predict O_3 peaks. These include multiple linear regression[11–13], generalized additive models, classification and regression tree analysis[11], and application of principal component analysis and clustering technique[14,15]. However, the O_3 formation process involving precursor emissions, atmospheric transport and mixing, and a complex system of photochemical reactions, is extremely nonlinear and non-stationary. None of the traditional statistic models are sufficiently dynamic to capture the rapid fluctuations in the O_3 time series. Therefore, all these models appear to have some difficulty in forecasting high O_3 events.

Artificial neural network (ANN) is another statistical approach which is frequently used in atmospheric research[16]. ANNs, which can be trained to approximate virtually any smooth, measurable function, have become popular in atmospheric science and have produced promising results. In particular, the use of the neural networks in air quality modelling has been shown to give acceptable results for atmospheric pollution forecasting of pollutants such as O_3[17–21], SO_2[22], PM_{10}[23,24], and $PM_{2.5}$[25].

Bangkok, where the highest O_3 level is recorded in the period from January to April[26], has been the target of deterministic models for simulation of O_3 episode for assessment of potential impacts of different management strategies on air quality[27] and scenarios study[28]. So far, no prediction tools have been applied to forecasting ground level O_3 in the city. This study therefore has been carried out to assess the applicability of ANN for O_3 prediction in Bangkok. The feed forward backpropagation neural network, i.e., the multi-layer perceptron (MLP), was used to develop a model for prediction of daily maximum 1-h O_3 levels in the city. The performance of the model was compared to the results from the multivariate linear regression (LR) model, which is applied to the same data set. The daily maximum 1-h O_3 concentration is of interest as Thailand is still subject to the 1-h O_3 standard (100 ppb).

METHOD

Input data preparation

There are 13 automatic ambient air quality stations in Bangkok operated by the Thailand Pollution Control Department (PCD). These stations are equipped to monitor carbon monoxide, total suspended solid (TSP), particulate matter (PM_{10}), sulphur dioxide, nitrogen monoxide, nitrogen dioxide, ozone, methane, and non-methane hydrocarbons (NMHC). The monitoring stations in Bangkok are divided into two categories; the general ambient air quality monitoring stations that are located 50–100 m from main roads and the curbside street-level ambient air quality monitoring stations that are 2–5 m from main roads. The monitoring data for Bangkok were collected on hourly basis covering the period of January 2000 to August 2003 from the PCD when O_3 monitoring data was only available at 11 stations (out of 13 stations) including 10 ambient stations and 1 roadside station.

Analysis of O_3 pollution during the selected period shows that the curbside stations, as expected, are characterized by a lower frequency of O_3 exceeding the Thailand hourly Ambient Air Quality Standard (AAQS) of 100 ppb than the ambient stations. Highest maximum 1-h O_3 levels exceeding the AAQS were recorded at ambient stations at a distance from the city centre. The results of the analysis also indicated that high O_3 pollution in Bangkok occurred mainly in the period from January to April (winter and local summer) and the lowest pollution was during the mid-rainy season in August, which is similar to the results of a previous study[26]. Examination of the diurnal distribution frequency of maximum 1-h O_3 levels over all the monitoring stations shows the highest daily O_3 concentrations in Bangkok occur in the period from 13:00–15:00. During the dry season period covered by our study, the daily O_3 level is observed to sharply increase from 10:00 to 11:00 and normally reach an already high values at 11:00 but the maxima are observed only in the afternoon, at 13:00 or later[26].

The O_3 data used in this study are the highest values of daily maximum 1-h concentrations observed among all the monitoring stations, i.e., the peak O_3 in Bangkok (one value per day over the entire the area covered by the monitoring network). Thus for each day during the study period from 1 January to 30 April for the years 2000–2003, the highest peak O_3 observed in the city was selected and used for modelling. Concentrations of O_3 precursors including NMHC, NO and NO_2 used for modelling are the averages of observed data of the respective pollutant over all monitoring stations in the study area between 6:00 and 9:00, which includes the morning rush hours of the day of interest. The input containing these average values produced a better model performance as compared to other trials with various combinations of individual hourly monitoring data. In fact, the maximum morning levels of the pollutants normally occur during this period but the hours of the maxima occurrence varied from one station to another[26]. In addition, taking the average value has helped to fill up the missing hourly data that occasionally occurred in the monitoring data series.

In this study meteorological data collected at the Bangkok Metropolis station, which is located in the city centre, were used. The meteorological data were obtained from the Thai Meteorological Department for the same period as the air quality monitoring data. The selected meteorological variables include the av-

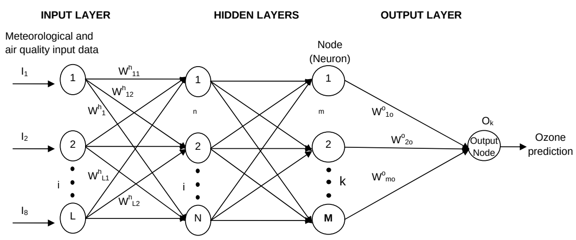

Fig. 1 Architecture of two hidden layer feed-forward neural network.

erage values from the observations in the morning hours (6:00 to 10:00) of wind speed (WS), relative humidity (RH) and global radiation (GRAD), and the daily maximum temperature ($T_{\max}$). Note that next day predicted meteorological variables are widely available for Bangkok and can be used instead of the observed ones. Accordingly, this would reduce the dependence of the observed data availability and increase the forecast horizon.

To remove the discontinuity of the wind direction (WD) angle at 360° the wind direction index (WDI) was used to represent the wind direction, which was calculated using

$$\text{WDI} = 1 + \sin\left(\text{WD} + \frac{\pi}{4}\right),$$

where WD is the wind direction (with 0° corresponding to the north). Thus, the WDI has a minimum of 0.07 for the southerly wind (180°) and a maximum of 1.96 when WD is 315°.

The total dataset used for the modelling included 481 lines (patterns – 1 per day) and 9 variables (NMHC, NO, NO_2, O_3, WS, WDI, RH, $T_{\max}$, and GRAD). The data set was randomly divided into two subsets: the first subset of 361 patterns (75%) was used for training the MLP model and for development of the regression model, and the remaining 120 patterns formed the second data subset (25% of the data) that was used for the model validation.

Artificial neural networks

Artificial neural networks (ANNs) are computer programs designed to emulate biological neural networks (such as the human brain) in terms of learning and pattern recognition. ANNs have been under development for many years in a variety of disciplines to derive meaning from complicated data and to make predictions. The most popular ANN is feed-forward back-propagation, multi-layer perceptron (MLP) neural network, which has found applications in atmospheric science [16].

The major building block for any ANN architecture is the processing element or neuron. An ANN generally consists of three or more layers: an input layer, one or more hidden layers, and an output layer. This study uses a four-layer feed-forward back-propagation neural network architecture as shown in Fig. 1. The input neurons receive data from external sources to the system, the hidden neurons receive signals from all of the neurons in the preceding layer, and the output neurons send information out of the system. These neurons are connected together by lines of communication called connections. Associated with each connected pair of neurons is an adjustable value or weight.

In this study we selected the feed-forward back-propagation MLP to develop the ANN model. Eight of the variables listed above (nine variables minus O_3) were used as inputs and the output from the MLP model was the daily maximum 1-h O_3 level over Bangkok (one value per day in the study area). All input variables were normalized to provide values between 0.05 and 0.95 using

$$O'_i = \frac{0.9(O_i - O_{i,\min})}{O_{i,\max} - O_{i,\min}} + 0.05,$$

where O'_i is a transformed observed value, O_i the actual observed value, $O_{i,\min}$ and $O_{i,\max}$ are the minimum and maximum values among all observed values over Bangkok. Normalization of input data was performed for two reasons: to provide commensurate data range so that the models were not dominated by any variable that happened to be expressed in large numbers, and to avoid the asymptotes of the sigmoid function. Once the best network is found, all the transformed data were converted back into their original values using

$$O_i = \frac{(O_{i,\max} - O_{i,\min})(O'_i - 0.05)}{0.9} + O_{i,\min}.$$

In the training process, the number of hidden layers and hidden nodes, and connection weights between neurons of the MLP network were determined by an iterative process in training stage with the training subset (361 patterns in this study) until the training error, measured by a set of performance indicators, is below the acceptable level. The initial values of the weights were randomly selected and they could be both negative and positive values. The activation function used in the hidden and output layers was determined by the required degree of accuracy of the problem under study. In this study, the learning algorithm used was Levenberg-Marquardt back-propagation of the MATLAB Neural Network Toolbox. The activation function selected for the layers was logistic sigmoid for hidden layers and linear for the output layer. The number of hidden layers and hidden neurons (nodes) were increased systematically, checking each time if the prepared neural network produced the stable performance error in the performance plot until the best MLP was found. The trained MLP network model was then validated with the second data subset of 120 patterns. The resulting predictions were then compared with the observed data, and performance indicators were determined.

Performance indicators

The performance indicators of the models were determined to provide a numerical description of the accuracy of model predictions, and to compare the performance among the models. A range of performance indices are generally used for model performance evaluation[29–31]. The general statistical measures considered here include the mean absolute error (MAE), the root mean square error (RMSE), the coefficient of determination (R^2), and the relative measure of error called the index of agreement (d)[32,33]:

$$\text{MAE} = \frac{1}{n}\sum_{i=1}^{n} |P_i - O_i|$$

$$\text{RMSE} = \sqrt{\frac{1}{n}\sum_{i=1}^{n} (P_i - O_i)^2}$$

$$R^2 = \frac{\sum_{i=1}^{n} (P_i - \bar{O})^2}{\sum_{i=1}^{n} (O_i - \bar{O})^2}$$

$$d = 1 - \frac{\sum_{i=1}^{n} (P_i - O_i)^2}{\sum_{i=1}^{n} (|P_i - \bar{O}| + |O_i - \bar{O}|)^2}$$

where n is the number of data points, P_i the predicted data point, and $\bar{O}$ is the average of the observed data.

We also calculated one statistical measure recommended for urban-scale O_3 model[29], the unpaired peak prediction accuracy (UPA) which is given by UPA $= (P_i - O_i)/O_i$ and is meant to evaluate the model's ability to reproduce the highest observed concentration anywhere in the study area. It is therefore considered to be suitable for this study as our concern is the maximum O_3 in Bangkok on each day during the study period.

Thus, for each day of the study period, there is one pair of observation-prediction maximum O_3 levels. We used a cutoff value of 60 ppb O_3 for observed O_3. Hence only the observation-prediction pairs with observed O_3 above 60 ppb were used for UPA determination. The US EPA (1991) guidelines give a recommended UPA range of ± 20%. Table 1 shows the performance evaluation results of the models.

Table 1 Performance indicators for the developed models.

Indicators	MLP		LR	
	Training	Testing	Training	Testing
MAE (ppb)	8.6	10.3	16.9	16.4
RMSE (ppb)	11.8	13.5	21.4	22.4
R^2	0.89	0.85	0.39	0.34
d	0.92	0.89	0.74	0.68
UPA (%)	−26 to 29	−24 to 23	−58 to 70	−63 to 56

Multivariate regression model

The accuracy of the developed MLP network was also compared to that of a LR model with ordinary least squares developed based on the same data set. The LR model between the eight input variables and the output (peak O_3) was performed using a stepwise regression analysis on the first data subset (training subset for MLP) to determine the regression coefficients.

The preliminary regression model has the general form:

$$Y = \beta_0 + \beta_1 X_1 + \beta_2 X_2 + \beta_3 X_3 + \cdots + \beta_k X_k + \varepsilon,$$

where Y stands for the predictand variable Y (e.g., daily maximum O_3), β_i, $i = 0, 1, 2, \ldots k$, are the regression coefficients, X_i is a set of k predictor variables X with corresponding β coefficients, and ε is the residual error.

Regressions were performed using the SPSS software package. The developed regression model was also tested for performance using the 2nd data subset used for the MLP validation.

RESULTS AND DISCUSSION

Linear regression model

The stepwise regression procedure on the first data subset (the same subset used for training the MLP) showed that NO_2, $T_{\max}$, WS, RH, NMHC, and WDI are all important for prediction of daily maximum 1-h O_3 in Bangkok (Table 2). The best single variable among the eight independent variables is NO_2 as expected since NO_x is an important O_3 precursor. The second best single variable was $T_{\max}$, which is most probably due to the fact that the photochemical reaction rates are temperature dependent[34].

The LR model $O_3 = 0.650\,NO_2 + 2.983\,T_{\max} - 12.738\,\text{WS} - 0.806\,\text{RH} + 6.367\,\text{NMHC} - 4.26\,\text{WDI} +$

Table 2 Stepwise regression results.

Steps	Set of variables	R^2
1	NO_2	0.200
2	NO_2, $T_{\max}$	0.273
3	NO_2, $T_{\max}$, WS	0.315
4	NO_2, $T_{\max}$, WS, RH	0.351
5	NO_2, $T_{\max}$, WS, RH, NMHC	0.371
6	NO_2, $T_{\max}$, WS, RH, NMHC, WDI	0.396

6.121 was found to give the best fit, with a MAE of 16.9 ppb, a RMSE of 21.4 ppb, a R^2 of 0.39, and a d of 0.74 (Table 1). This model was then applied to the testing subset of data (the same as for MLP) for prediction. Scatter plots of the actual monitoring O_3 concentrations versus the predicted values by this model for the training and testing subsets of data are given in Fig. 2. The time series of the predicted results and the observed O_3 concentrations for the testing dataset is shown in Fig. 3. This simple statistical model, as expected, underestimates the peak O_3 values at the beginning and at the end of the data series (Fig. 3). Better prediction is recorded in the middle of the data series, from the 20th to 70th data points. In general, the performance of this simple statistical model is considered to be poor.

Artificial neural network model

The iterative process in the training stage found that the architecture of the best MLP network should be 8-10-14-1, i.e., it should have the input layer containing 8 neurons, the first hidden layer containing 10 neurons, the second hidden layer containing 14 neurons, and the output layer with 1 neuron. The scatter plots of predicted and observed O_3 concentrations for the training and testing datasets are illustrated in Fig. 4. The MAE and the RMSE for the training dataset are 8.6 and 11.8 ppbv, respectively. The corresponding errors for the testing dataset were 10.3 and 13.5 ppbv, respectively. The time series of predicted versus observed O_3 concentrations for the two datasets (Fig. 5 and Fig. 6) show that the predicted values are in a good agreement with the observed ones. In particular, the peak O_3 values in the time series shown in Fig. 6 have been captured satisfactorily by this MLP model which is better than the simple LR model discussed above.

Comparative analysis of the performance of the models

The MAE and the RMSE values of the MLP predicted values are lower than those from the LR model for both training and validation data subsets (Table 1).

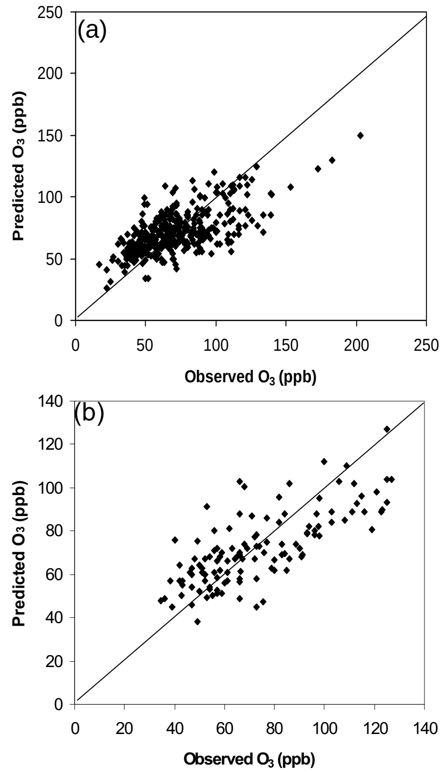

Fig. 2 Scatter plots of observed versus predicted O_3 levels by the regression model: (a) training dataset (b) testing dataset.

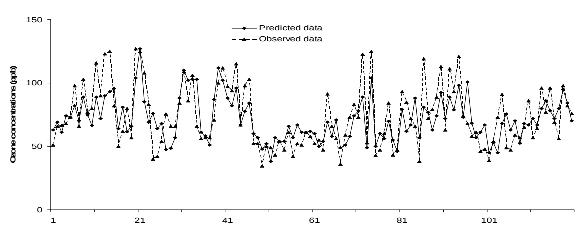

Fig. 3 Observed versus predicted O_3 levels by the LR model for the testing dataset.

The peak values in the O_3 data series were underestimated by LR (Fig. 7). The reason for the underestimation is that the fitting of regression coefficients is solved using a least-squares method, which minimizes the sum of squared errors[37]. Therefore, the LR does not capture the extreme values. The regression analysis process aims at modelling the 'average' behaviour for the predictand (output) variable, whereas with regards to air quality standards, the prediction of extreme O_3 levels is much more important from the health perspective. Due to the high nonlinearity of the O_3 photochemical formation process and the complex

Table 3 Overview of selected studies on prediction daily maximum O_3 using ANNs.

Reference	Location	Data set	MAE	RMSE	R^2
Comrie[19]	USA, 8 cities	1991–1995	7.01–13.46	8.82–17.53	0.37–0.69
Cobourn et al[35]	USA, 7 sites	1998–1999	11.6–13.4	14.4–16.9	NA
Chaloulakou et al[24]	Athens, 4 sites	1992–1999	17.3–32.6	12.6–21.1	0.45–0.76
Zolghadri et al[36]	Bordeaux, France	1998–2001	11.6	NA	NA
This study	Bangkok	2000–2003	8.6–10.3	11.8–13.5	0.85–0.89

NA, not available

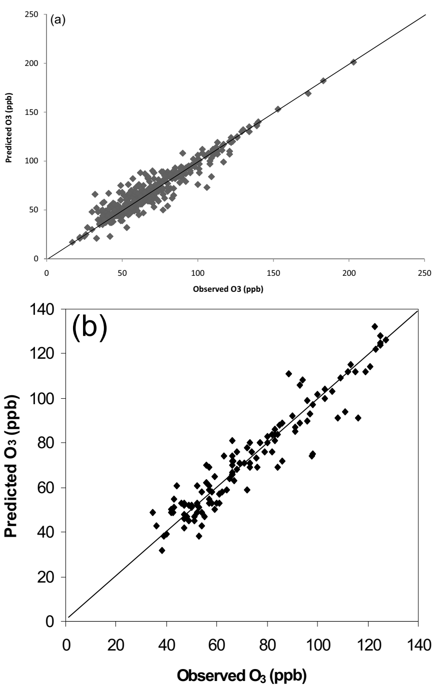

Fig. 4 Scatter plots of observed versus MLP model predicted values: (a) training dataset (b) testing dataset.

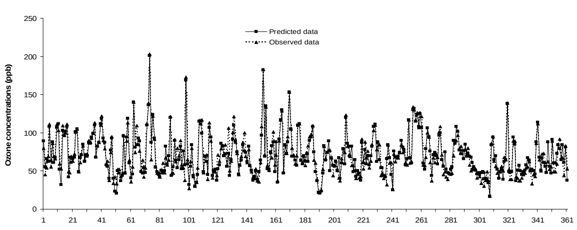

Fig. 5 Observed versus predicted O_3 by the MLP model for the training dataset.

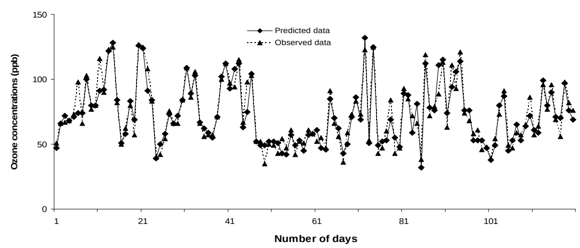

Fig. 6 Observed versus predicted O_3 by the MLP model for the testing dataset.

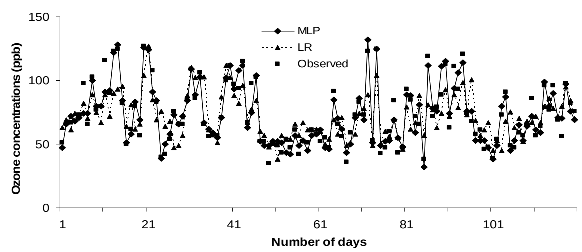

Fig. 7 Comparison of O_3 prediction models on the testing dataset.

interactions between meteorological variables and O_3, the MLP gives better predictions for the maximum daily 1-h O_3 values that exceed the Thailand AAQS of 100 ppbv.

In the present study, UPA ranged from −26 to 29% and from −24 to 23% for training and validation data subsets of the MLP modelling, respectively. The corresponding values for the LR model ranged from −58 to 70% and from −63 to 56%, respectively. For the MLP model, there were 21 cases (5.8%) in the training dataset and 6 cases (5%) in the validation dataset which were larger than ±20%. Thus, the majority of the predictions by MLP for both data subsets met the US EPA criterion. For the LR model, the percentages of the predicted values that did not meet this UPA criterion are higher, i.e. 90 pairs (25.0%) and 33 pairs (27.5%), respectively, were larger than ±20%. Thus, considering this US EPA UPA criterion, the performance of the MLP model is largely satisfactory and almost comparable to those 3D models mentioned above. This further suggests that the MLP model is suitable for air pollution predictions when there is no accurate emission data available. It is worth mentioning that the lack of an accurate emission database is one of the obstacles for the application of sophisticated deterministic models in many places. Previous simulations of O_3 episodes in the Bangkok metropolitan region using 3D deterministic models, Variable Grid Urban Airshed Model (UAM-V) and CHIMERE[27,28], based on the PCD emission database, produced the O_3 prediction with a UPA that generally did not meet the US EPA criteria (within ±20%). These studies suggested that the PCD emission database should be further revised. In fact, when the 3D models were run with a hypothesized/modified emission input data the performance in meeting the UPA criteria was improved.

Based on MAE, RMSE and R^2 (Table 3), the performance of our MLP model are similar to other published studies of ANN O_3 prediction models worldwide but with a higher R^2. It is interesting to note a similarity in the performance of the ANN models among reviewed studies, although the test cases were applied to different urban environments, different meteorological conditions and at different time periods.

The MLP model developed in this study has been specifically constructed for Bangkok based on the data of the period with the highest O_3, 1 January–30 April. This selection of the data period, however, largely excluded effects of seasonal factors in the model formulation and performance. The model has not been tested on the additional data sets, which is required to check the model generalization on unseen data[16,30]. Thus, model testing should be conducted for a large enough dataset before application for operational purpose. In addition, a new model may be required for the wet season when O_3 is generally low.

CONCLUSIONS

The developed MLP model with 2 hidden layers performs satisfactorily in prediction of the daily maximum 1-h O_3 concentrations in Bangkok. The model performance is significantly better than the LR model obtained by the stepwise regression, especially for the peak O_3 concentrations. The MLP model is a simple model, which can provide relatively reliable estimates of maximum daily O_3 based on only limited ambient air monitoring data. The ANN approach can be used to develop models for prediction of air pollution in developing countries where a common lack of the accurate emission data prevents the application of more sophisticated dispersion models. The prediction models developed in this study use the input consisting of the air quality and meteorological variables measured in the morning of the prediction day. Next day predicted meteorological variables can be used to reduce the dependence of the observed data availability and increase the forecast horizon. Thus, the model can provide the predicted O_3 level 4–5 h in advance which would provide enough time for warning as daily maxima in Bangkok occur in the afternoon. A longer forecast horizon, however, is desirable for effective warning of high O_3 concentrations. This calls for future research to focus on the development of ANN models to provide, for example, the next day maximum O_3 forecast. Other variables, such as the previous day O_3 maxima, can be used to improve the model performance. Intensive testing with a large dataset is still required before the developed MLP model can be applied for the operational purpose. Other model performance indices that are applicable when the model is applied to assess the predicted O_3 levels against the ambient air quality standard such as verification statistics for category forecasts[30] should be considered. Furthermore, the MLP model can be used in combination with other forecasting methods to further ensure the applicability over different periods of the year.

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