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MODELLING PARTICULATE MATTER (PM_{10}) CONCENTRATION IN INDUSTRIALIZED AREA: A COMPARATIVE STUDY OF LINEAR AND NONLINEAR ALGORITHMS

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ABSTRACT

Particulate matter is a critical air pollutant in Malaysia as it is the utmost dominant pollutant, especially in industrial and urban areas. The development of a robust model for PM_{10} concentration forecasting provides invaluable information for local authorities to take precautionary measures and implement significant actions to improve air pollution status. This study aims to develop and assess the linear (Multiple Linear Regression, MLR) and nonlinear (Multilayer Perceptron, MLP) models forecasting capability in industrial area of Pasir Gudang, Johor. Daily observations of PM_{10} concentration, meteorological factors (wind speed, ambient temperature and relative humidity) and gaseous pollutants (SO_2, NO_2 and CO) from the year 2007-2014 were used in this study. Results showed that MLP model was able to explain 68.7% (R^2 = 0.687) variance in the data compared to MLR model with 52.7% (R^2 = 0.527). Overall, the MLP model able to increase the accuracy of forecasting by 29.9% and reducing the error by 69.3% with respect to MLR model. Thus, it is proven that nonlinear model has high ability in virtually representing the complexity and nonlinearity of PM_{10} in the atmosphere without any prior assumptions, unlike the linear model.

Keywords: air quality, forecasting, industrial area, nonlinear.

INTRODUCTION

Air pollution is explaining as the pollutants start by the human, indirectly or directly, into the air which brings about unfavourable impacts to human particularly in wellness viewpoint and the environment [1]. Anthropogenic sources and natural sources are two major sources that contribute to the pollution in the ambient air [2]. It is regularly found that the PM_{10} concentration is the most elevated contrasted with other pollutants [3]-[4]. An adverse effect on human health by air pollutants [5], especially particulate matter, has received extensive attention from researchers [6]-[8]. Among the other five major PM_{10} emissions listed by DOE [3], emission from industrial activity is the prominent sources in Malaysia [9]-[10]. Thus, forecasting of PM_{10} concentration at industrial area as the main strategy in air quality management is very crucial.

The characters of PM_{10} in the atmosphere are influenced by several meteorological factors and gaseous pollutants which control the dispersion, formation and transportation of PM_{10} [11]. This triggers to the complexity and nonlinearity of PM_{10} in the air. In representing the real-world situation of the nonlinear character of PM_{10} in the atmosphere without creating any assumption (Al-Alawi *et al.*, 2008), unlike the traditional (linear) model, this study utilizes the Computational Intelligence (CI) technique known as Artificial Neural Network (ANN). ANN is the analogy on how the human brain is functioning [12] in memorizing, thinking and delivers information. Multilayer Perceptron (MLP) model is a member of ANN that flexible and able in approximating any smooth differentiable function, along these lines, it is relevant particularly while showing complex nonlinear procedures [13]. Therefore, by taking all these points into consideration, this study was conducted in developing the nonlinear model for PM_{10} concentration forecasting at industrialized area using ANN model, in comparison of the traditional model of Multiple Linear Regression (MLR) model.

MATERIALS AND METHODS

SITE SELECTION

Pasir Gudang, an industrial area in the State of Johor is located in the southern region of Peninsular Malaysia. There were several industries that played an important role in the economic development for Pasir Gudang including petrochemical, oleochemical, oil and gas, and plastic products [14]. The location of Air Quality Monitoring Station (AQMS) for Pasir Gudang is precisely located at SMK Pasir Gudang 2 (1°28'14.76" N; 103°53'43.87" E) (Figure-1).

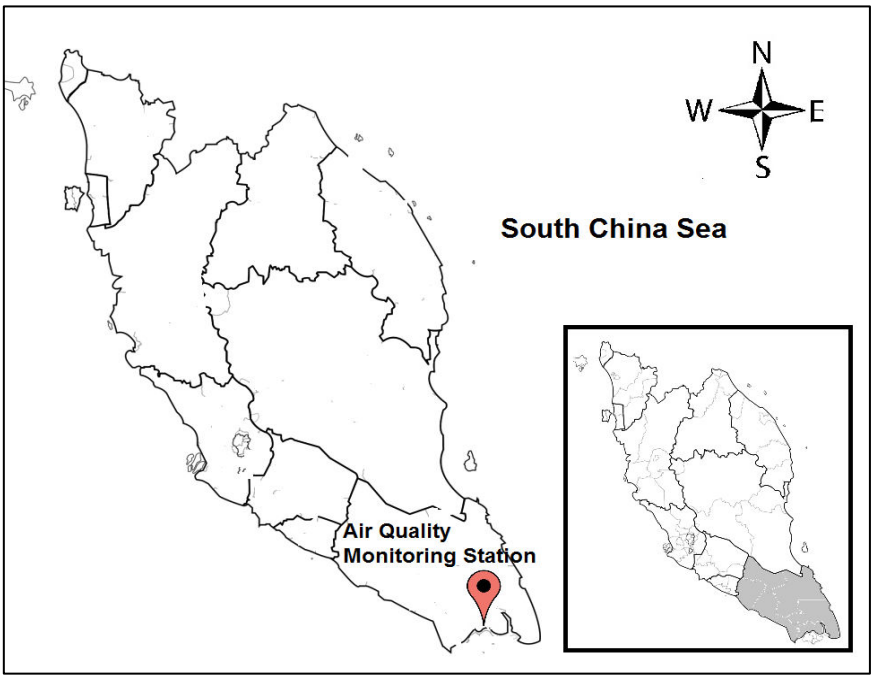

Figure-1. Location of study area.

ACQUISITION AND PRE-PROCESSING OF DATA

This study was conducted based on 8 years data covered from the year 2007-2014, obtained from Malaysian Department of Environment. The data at first is given in hourly measurement that already contains the daily concentration PM_{10} in the unit of μg/m^3. Meteorological variables for instance the ambient temperature (^{0}C), wind speed (ms^{-1}), and relative humidity (%) and other gaseous pollutants such as nitrogen dioxide (NO_2, ppm), carbon monoxide (CO, ppm) and sulphur dioxide (SO_2, ppm) were considered in this study as the predictors (7 inputs). The data was divided into two parts; 70% (N=2045) for model training and 30%(N=877) for model testing, respectively [9] [15]. PM_{10}, relative humidity, wind speed, ambient temperature and gaseous pollutants (CO, NO_2, and SO_2) were known as input parameters (7 inputs) and the output parameter is the next day of PM_{10} concentration.

The measurement error and breakdown of instruments might result in missing values [16]. Thus, it can cause bias due to systematic differences between measured and predicted data. The missing values from the data obtained were calculated at which the number of missing data per row is divided by the total number of data per year and multiplied by 100%. New values were inserted within the set of data by an interpolation technique and linear interpolation method was used in this study to impute the missing values of the acquisition data as the calculated data has less than 25% of missing per year. In SPSS version 23, the last and the first available values of that particular missing data were used for insertion of missing values by means of linear interpolation [17]. Linear interpolation connects two data points with straight line [16]. By adaptation concepts of algebra, the equation for linear interpolation is given as:

$$f(x) = f(x_0) + \frac{f(x_1)-f(x_0)}{x_1-x_0}(x - x_0) \quad (1)$$

x = independent value
x_1 and x_0 = known values of the independent variable
$f(x)$ = value of dependent variable for a value of the independent variable

Normalization is required due to the input parameters of the model that have different units. The range of 0 to 1 is generated as the result of normalization process with min-max technique [0 1] [18]-[20] and it helps for execution of better outputs for ANN model [21]. The normalization of data is accompanying following transformation [22]:

$$z_i = \frac{x_i - min(x)}{max(x) - min(x)} \quad (2)$$

where, $x = (x_1, \dots, x_n)$ and z_i is the ith normalized data.

MULTIPLE LINEAR REGRESSION (MLR) MODEL

Multiple linear regression endeavours to demonstrate the relation between at least two independent variables and a dependent variable by fitting a linear information. This relationship is expressed in a mathematical equation. Generally, the equation of MLR is as follows:

$$y = b_0 + \sum_{i=1}^{n} b_i X_i + \varepsilon \quad (3)$$

Where b_i are the regression coefficients, X_i are the independent variables and ε is stochastic error

associated with the regression. The assumptions of MLR include the normal distribution, uncorrelated and constant variance in terms of its residuals. The method used in obtaining the model was stepwise multiple linear regression. The forecasting models are based on 95% confidence interval [23].

Multicollinearity assumption will be verified by Variance Inflation Factor (VIF) accompanied with the regression output, where as long as the average VIF is under 10 the conducted regression should be fine, signifying multicollinearity doesn't exist among independent variables [24]. The VIF is given by:

$$VIF_I = \frac{1}{1-R_i^2} \quad (4)$$

Where, VIF_i is the variance inflation factor associated with the ith predictor and R_i^2 is the multiple coefficient of determination in a regression of the ith predictor on all other predictors.

Durbin-Watson (D-W) Test measures the autocorrelation. Autocorrelation basically uncovers the ability of PM_{10} concentration from current day to forecast PM_{10} concentration on the following day. D-W test values will be in the range of 0 and 4, in case the determined value of 2 implies it is uncorrelated among residuals data [10]. The D-W is given by:

$$d = \frac{\sum_{i=1}^{n}(e_i - e_{i-1})^2}{\sum_{i=1}^{n} e_i^2} \quad (5)$$

Where n is number of observations, $e_i = y_i - \overline{y}_i$ (y_i = observed values and $\overline{y}_i$ is the predicted values)

MULTILAYER PERCEPTRON (MLP) MODEL

Multilayer Perceptron (MLP) is a type of feed forward neural network which has the ability in developing the nonlinear models with high complexity, make it preferred in air pollution forecasting [25]. The MLP starts when the interested input parameters are feed into the network. These input parameters provide input signals and these signals are sent in the network starting from input layer to hidden layer and hidden layer to output layer. The scaled input vector which introducing by neurons in the input layer is multiplied with weights, which a real number quantity. The neuron in hidden layer sums up this information, including bias.

$$y_o = \sum_{i=1}^{n} w_i x_i + b \quad (6)$$

This weighted sum information is still in its linear model. The non-linearity of information or model occurs when it is passing through the activation or transfer function.

$$f(x) = \frac{1}{1+e^{-x}} \quad (7)$$

Then,

$$y_o = f\left[\sum_{i=1}^{n} w_i x_i + b\right] \quad (8)$$

Where, y_o is the output, W_i is weight vector, X_i is the scaled input vector, b is the bias, f is the transfer function and x represents the total sum of weighted inputs.

Once the error signal is computed, the process of model fitting is ended. The difference between target and output is used to compute the error signal in the model which corresponds to the input [26].

Mathematically, the equation of MLP with several numbers of neurons is given as;

$$y_o = f\left[\sum WO_{kj}\left(\sum_{i=1}^{n} WI_{ij} x_i + b_1\right) + b_2\right] \quad (9)$$

Where, WI_{ij} and WO_{kj} are the weights of input and output layers, respectively, b_1 and b_2 are biased in input and output layer, respectively.

The determination of neuron numbers in the hidden layer is the most critical part as there is a strong impact on the output in which determines by the neuron numbers in the hidden layer. Under-fitting will occur if the number of the neuron is too few and too many of neurons contribute to over-fitting, thus the determination of an optimum number of the neuron is really important [27]. Trial and error method was employed for the determination of neuron numbers in the hidden layer. It was stated that the neuron numbers should not be twice the numbers of input variables [24].

The MLP model was trained by using Lavenberg-Marquardt (LM) training algorithms in treat the weight and bias. The LM approach is analteration of the classic Newton algorithm for finding the best solution for minimizing the problem. It uses an approximation to the Hessian matrix in the following Newton-like weight update:

$$x_{k+1} = x_k - [J^T J + \mu I]^{-1} J^T e \quad (10)$$

$x =$ Weight of neural networks
$J =$ Jacobian matrix of the performance criteria to be minimized
$\mu =$ Scalar that controls the learning process
$e =$ The residual error factor

Equation 10 is just Newton's method when the scalar μ is zero by using the approximate Hessian matrix. Equation 10 will becomes gradient drop with a small step size when μ is large. Newton's method is quicker and more precise near to least possible of any error. Great computational and memory requirements are the advantages of LM thus it is suitable to be used in small networks [28].

The initialization of the model was trained with 1000 epochs and the model was run 10 times to get the average of the value. The learning rate is set as 0.05 to train the optimum configuration of MLP network and to improve the performance of the model [29]. The hyperbolic tangent sigmoid (tansig) and linear transfer function (purelin) were used as activation functions in the

hidden and output layer, respectively. MATLAB R2015a was used for models training. Figure-2 shows the architecture of neural network model that consists of three layers which are input, hidden and output layer.

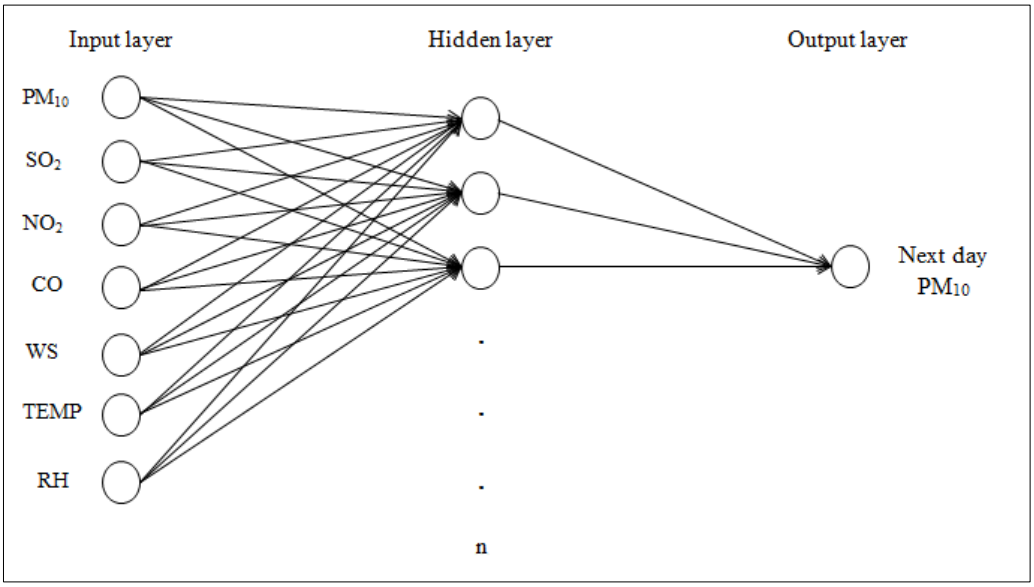

Figure-2. Architecture of ANN model.

PERFORMANCE INDICATORS

Five performance indicators (PI) were used in determining the best-fitted models in this study. Based on PI, there are two things that are taken into account, namely accuracy measure and error measure. The accuracy measure indicates that the best-fitted model is when the value is closer to 1 while error measure indicates the best model when the evaluated value is closer to 0 [2];

a) Root Mean Square Error

$$RMSE = \left(\frac{1}{n}\sum_{i=1}^{n}[P_i - O_i]^2\right)^{\frac{1}{2}} \quad (11)$$

b) Normalized Absolute Error

$$NAE = \frac{\sum_{i=1}^{n}|P_i - O_i|}{\sum_{i=1}^{n} O_i} \quad (12)$$

c) Mean Absolute Error

$$MAE = \frac{\sum_{i=1}^{n}|O_i - P_i|}{n} \quad (13)$$

d) Correlation Coefficient

$$R^2 = \left(\frac{\sum_{i=1}^{n}(P_i - \bar{P})(O_i - \bar{O})}{n.S_{pred}.S_{obs}}\right)^2 \quad (14)$$

e) Index of Agreement (IA)

$$IA = \left[\frac{\sum_{i=1}^{n}(P - O_i)^2}{\sum_{i=1}^{n}(|P_i - \bar{O}| + |O_i - \bar{O}|^2}\right] \quad (15)$$

$n =$ total number of data
$P_i =$ predicted values
$O_i =$ observed values
$\bar{P} =$ mean of predicted values
$\bar{O} =$ mean of observed values
$S_{pred} =$ standard deviation of predicted values
$S_{obs} =$ standard deviation of the observed values

RESULTS AND DISCUSSIONS

TREND OF PM_{10} CONCENTRATION

The trend of PM_{10} concentration over the study period was assembled through Figure-3. It was noted that the highest annual mean was recorded as 58.32 µg/m^3 in 2009, while the lowest annual mean of PM_{10} concentration was in 2007 with 44.72 µg/m^3. Daily maximum PM_{10} concentration was observed in the year 2013 with 461.25 µg/m^3, while the minimum concentration is 11.48 µg/m^3 in the same year. In the year 2015, the New Ambient Air Quality Standard (NAAQS) had been developed by Malaysian Department of Environment (DOE) as a reference for determination of air quality in Malaysia. The limit was set up to not exceeded 50 µg/m^{3}for annual (yearly) average and 150 µg/m^3 indicates the daily average for PM_{10} concentration [30]. This station exceeds the daily limit in year 2008 (154.5 µg/m^3), 2009 (157.5 µg/m^3), 2010 (153.8 µg/m^3) and 2013 (461.25 µg/m^3) and exceeds the yearly limit in year 2008, 2009, 2012, 2013 with the reading of 53.24 µg/m^3, 58.32 µg/m^3, 51.98 µg/m^3 and 51.03 µg/m^3, respectively. Previous studies had successfully aligned, the increasing of PM_{10} concentration in Peninsular Malaysia was related to the trans-boundary smoke resulted from the forest fire that came from Sumatera region of Indonesia [2] [31], occurred especially during the dry season months (May to September).

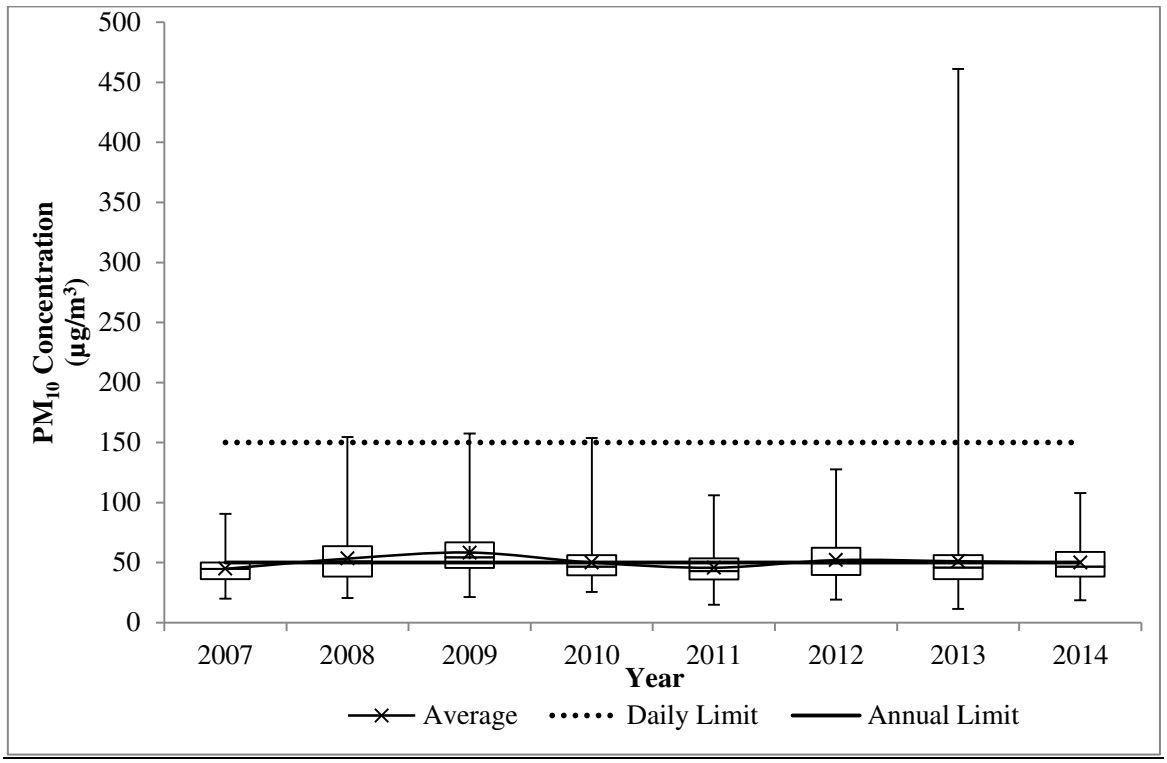

Figure-3. Trend of PM_{10} concentration at pasir gudang station.

DEVELOPMENT OF MLR MODEL

The investigation of the air quality and meteorological information is proceeded by applying Multiple Linear Regression (MLR) with the extra point of creating viable operational models for PM_{10} concentration forecasting. The MLR model was developed and the model's summary is depicted in Table-1.

Table-1. Summary of MLR model for PM_{10} forecasting.

Model	R^2	Range of VIF	Durbin Watson Statistics
$PM_{10,t+1}$ = 0.508 (PM_{10}) + 0.226(NO_2) - 0.094(Relative Humidity) + 0.106(CO) + 0.075(Ambient Temperature) + 0.105(Wind Speed) - 0.057(SO_2) - 0.029	0.527	1.209 - 4.203	2.001

The best MLR model was obtained with R^2 of 0.527, which means the developed model able to explain 52.7% the variance in data. The determined VIF values among the independent variables for MLR model was in the range of 1.209 - 4.203. The VIF values are less than 10, signifying multi-collinearity did not exist among independent variables [24]. The developed model did not face with autocorrelation problems as the determined Durbin Watson statistic was 2.001. It was found that the significant predictors were $PM_{10,}$ NO_2, relative humidity, CO, ambient temperature, wind speed and SO_2. The appropriateness of any developed statistical model can be measured through its residuals or error. The appropriateness of models was measured through residuals analysis. The model is considered containing error by showing the pattern from the results of residuals analysis. Figure-4(a) indicates histograms of the residuals of PM_{10} models. The residual analysis shows that the residuals are normally distributed with zero mean and constant variance. The plots of fitted values with residuals for PM_{10} model are shown in Figure-4(b), indicating that the residuals are uncorrelated because the residuals are contained in a horizontal band and hence obviously that variance is constant. The forecasted daily PM_{10} concentrations for the model was plotted in Figure-4(c) against the observed values to determine a goodness-of-fit of the models. The regression lines showing 95% confidence interval were also drawn. Most of the points fall in the range of 95% confidence interval. Lines A and C are the upper and lower 95% confidence limit for the regression model.

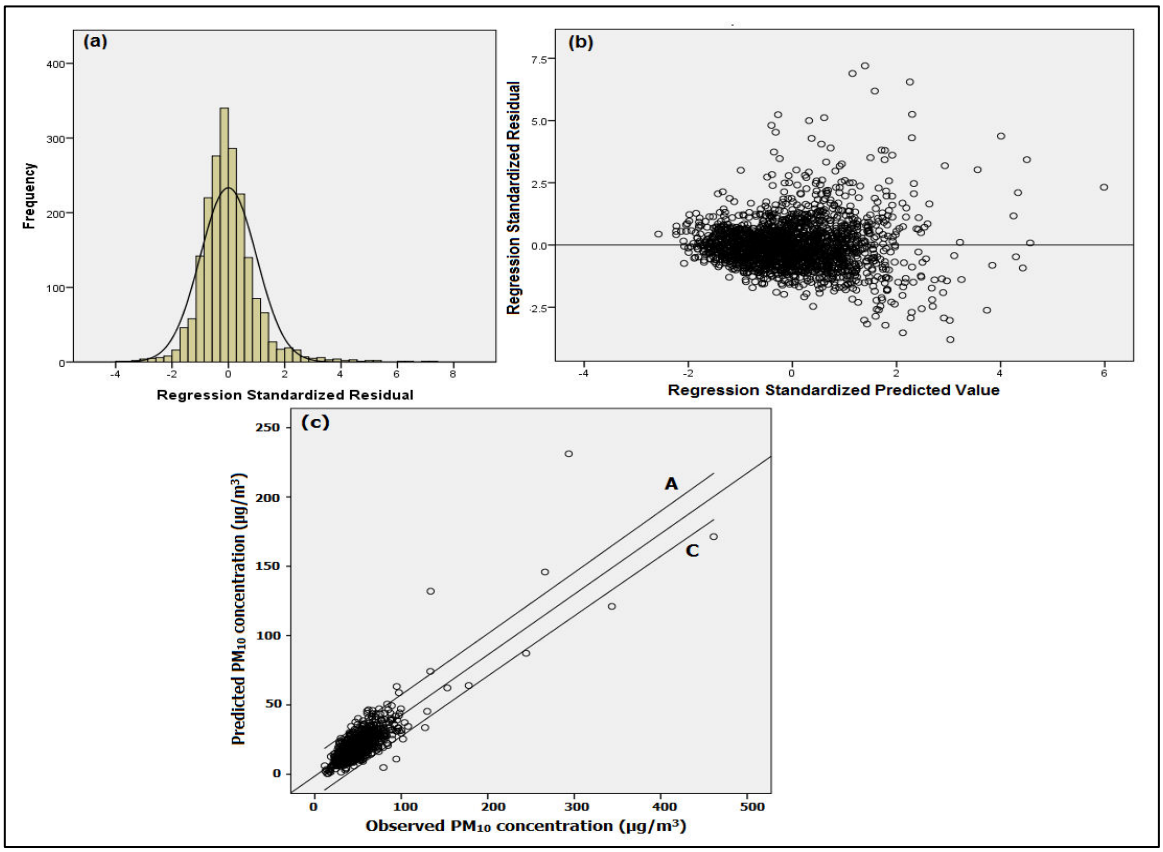

Figure-4. (a) Standardized residual analysis of PM_{10} (b) Testing assumption of variance and uncorrelated with mean equal to zero (c) Scatter plot for predicted PM_{10} concentration (µg/m^3) against observed PM_{10} concentration (µg/m$_3$). of PM_{10} concentration at Pasir Gudang Station.

TRAINING OF MLP MODELS

Table-2 shows the results using the different number of neurons for MLP model. It should be noted that the best results are marked with bold.

Table-2. The optimum number of neurons in hidden layer.

Number of neurons in hidden layer	RMSE (µg/m^3)	R^2
1	12.004	0.529
2	11.818	0.543
3	11.311	0.582
4	11.112	0.596
5	10.907	0.611
6	10.899	0.611
7	10.718	0.624
8	10.578	0.634
9	10.443	0.643
10	10.09	0.667
11	10.179	0.661
12	10.073	0.668
13	9.977	0.674
14	9.789	0.687
15	9.812	0.685

The selection of best model during training phase is strictly based on the R^2 and RMSE. The optimum number of neurons in hidden layer was 14. Although this study restricts the range of neurons in the hidden layer to be tested in training stage only up to 14, the addition of one neuron in the training stage (15 neurons) is important to pertinently prove the error to increase and the accuracy to decrease and the training were then terminated. The inputs that are feed into the MLP model able to explain 68.7% of the variance in data. The forecasted and observed of PM_{10} concentration was depicted in Figure-5.

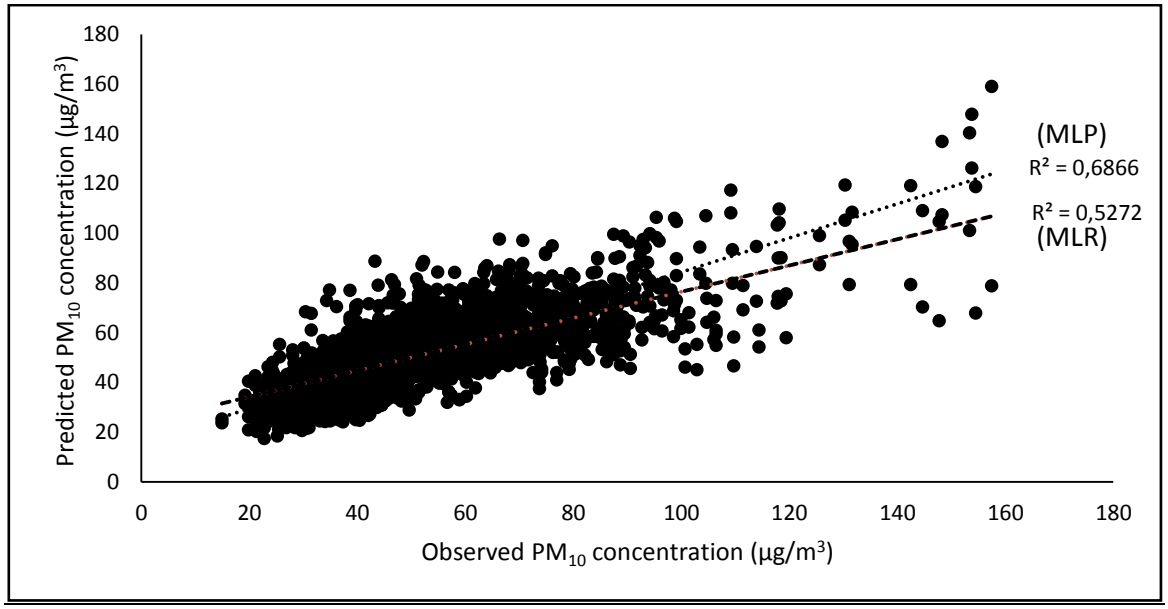

Figure-5. Predicted PM_{10} concentration (µg/m^3) against observed PM_{10} concentration (µg/m$_3$) during development of MLR model and training of MLP model.

MODELS EVALUATION AND SELECTION

Root Mean Square Error (RMSE), Normalized Absolute Error (NAE), Mean Absolute Error (MAE), Correlation Coefficient (R^2) and Index of Agreement (IA) were used in the process of evaluation and the selection of the best-fitted model. The results are presented in Table-3.

Table-3. The evaluation of performance indicators.

Multiple linear regression (MLR) **model**				
RMSE (µg/m^3)	**NAE**	**MAE** (µg/m^3)	**R^2**	**IA**
33.278	0.573	29.481	0.651	0.726
Multilayer Perceptron (MLP) Model				
RMSE (µg/m^3)	NAE	MAE (µg/m^3)	R^2	IA
11.389	0.166	8.519	0.839	0.950

The error measures such as RMSE, NAE and MAE show the best model if the evaluated values are near to zero, while R^2 and IA are known as accuracy measures which the values approaching one indicates a better model. It should be noted that the best results are marked with bold. The MLP indicates the low error measure of 11.389 µg/m^3 (RMSE) 0.166 (NAE) and 8.519 µg/m^3 and high accuracy measures of 0.839 (R^2) and 0.950 (IA) as compared to MLR model with 33.278 µg/m^3 (RMSE), 0.573 (NAE), 29.481 µg/m^3 (MAE), 0.651 (R^2) and 0.726 (IA).

This study looks at the profundity of the comparison between two models used in forecasting the next day of PM_{10} concentration. It was proven that the non-linear model, in particular, MLP is a better model in forecasting the next day of PM_{10} concentration rather than the linear model, which is MLR, since the statistical indices of the first were efficiently better contrasted with the ones of the reference model. The nonlinear model able in reducing the error of models by 69.3%. Interestingly, the nonlinear model proved to increase the accuracy of forecasting with 29.9%. This study was able proved that there was an improvement of nonlinear model in terms of reducing model's error and increasing model's accuracy for PM_{10} forecasting. This current result in this study is similar to the previous findings [24]-[25] [32]-[33]. Moreover, the performance of the MLP-ANN display is

extremely sensible and therefore it can be considered for operational utilize.

CONCLUSIONS

This study developed the linear and nonlinear algorithms as a comparative assessment in determining the better model for forecasting the next day PM_{10} concentration. Results indicated that nonlinear model outperforms the linear model as the evaluated performance indicators of nonlinear model exhibit lower error and higher accuracy, compared to the linear model. The ability to capture complexity, nonlinearity and without prior assumptions proved the robustness of the nonlinear model. This developed model is ready for operational usage which will provide invaluable information for local authorities to take precautionary measures and implement significant actions in improving air quality status.

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CONFLICT OF INTEREST

The authors declare no conflict of interests.

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