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Land use regression models coupled with meteorology to model spatial and temporal variability of NO_2 and PM_{10} in Changsha, China

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HIGHLIGHTS

- Daily NO_2 and PM_{10} concentrations were predicted based on land use and meteorology.
- Meteorological factors were introduced to improve temporal resolution of LUR model.
- A nonlinear relationship exists between meteorology and NO_2 and PM_{10}.

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ABSTRACT

Land use regression (LUR) models are widely used in epidemiological studies to assess exposure to air pollution. However, most of the existing LUR studies focus on estimating annual or monthly average concentration of air pollutants, with high spatial but low temporal resolution. In this paper, we combined LUR models with meteorological conditions to estimate daily nitrogen dioxide (NO_2) and particulate matter (PM_{10}) concentrations in the urban area of Changsha, China. Seventy-four sites for NO_2 and thirty-six sites for PM_{10} were selected to build LUR models. The LUR models explained 51% and 62% of spatial variability for NO_2 and PM_{10}. The most important spatial explanatory variables included major roads, residential land and public facilities land, indicating that the spatial distributions of NO_2 and PM_{10} are closely related to traffic conditions and human activities. Meteorological factors were introduced to model the temporal variability of NO_2 and PM_{10} by using meteorological factors regression (MFR) and back propagation neural network (BPNN) modeling techniques. Important temporal explanatory variables included temperature, wind speed, cloud cover and percentage of haze. Pearson's r values between predicted and measured concentrations were much higher in BPNN models than in MFR models. The results demonstrate that the BPNN models showed a better performance than the MFR models in modeling temporal variation of NO_2 and PM_{10}. The approach of modeling spatial and temporal variation of air pollutants using LUR models coupled with meteorological conditions has potential usefulness for exposure assessment, especially for medium or short term exposure, in health studies.

1. Introduction

Many studies have reported that exposure to air pollutants such as NO_2 and PM_{10} may cause acute or chronic health problems (HEI, 2010; Gonzales et al., 2012; Sally Liu et al., 2012). Accurate measures of personal exposures are of great importance for

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epidemiological studies on the health effects of air pollution. In recent years, many efforts to improve quantitative methods of assessing personal exposure have contributed to new approaches for exposure assessment in air pollution studies. These approaches including geostatistical interpolation (Jerrett et al., 2005; Janssen et al., 2008), dispersion models (Gauderman et al., 2005; Liu et al.,, 2007; Cyrys et al., 2005), and land use regression (LUR) models (Aguilera et al., 2008; Dons et al., 2013; Hoek et al., 2011). Interpolation of monitored concentrations does not adequately show true spatial variability because routine monitoring networks are usually not dense enough to reflect localized variation in pollutant concentrations. Dispersion models are extremely dependent on accurate and spatially resolved input data, especially for emissions. In recent studies, LUR models have been proven to be a valid and cost-effective approach for assessing exposure to air pollutants in epidemiological studies (Gulliver et al., 2013; Lee et al., 2014; Wu et al., 2011). Generally, LUR models utilize targeted pollutants concentrations measured at a limited number of sites (usually 20–100 sites)and land use characteristics derived from a geographic information system (GIS) to predict pollutants concentrations at unmeasured locations (Henderson et al., 2007; Saraswat et al., 2013). LUR models were first applied to an air pollution epidemiology study in Europe (Briggs et al., 1997). Since then, due to the increasing ability of GIS to provide land use data, this robust type of model is increasingly used in Asia and North America (Kashima et al., 2009; Mukerjee et al., 2009).

Quantification of the spatial and temporal variation of air pollutants could provide more accurate exposure assessment for epidemiological and other air pollution studies (Blanchard et al., 2014). In recent years, although the temporal resolution for LUR models has improved, most LUR model studies still focus on annual or seasonal average concentrations, with high spatial but low temporal resolution. Generally, annual or seasonal LUR models are enough to assess long term individual exposure in chronic epidemiological studies. However, for medium or short term exposure in acute epidemiological studies, annual or seasonal LUR models are insufficient to capture all of the variation. Recently, some studies have made efforts to improve temporal resolution of LUR model. The simplest method is to recalibrate existing LUR models with a continuous background monitoring station (Gan et al., 2011; Nethery et al., 2008). Another approach is to build several unique models in different time periods (Dons et al., 2013; Chen et al., 2012).

Urban areas possess complex spatial configurations, and these configurations are produced by cumulative change in land use (Wrenn and Sam, 2014). Usually, the land use predictor variables show a large spatial difference in a fixed period of time; in contrast, weather conditions vary from day to day, showing major change over time. Therefore, the land use and meteorological variables could be seen as spatial and temporal variables, respectively. The aim of this study is to build integrated daily models to model spatial and temporal variability of NO_2 and PM_{10} in the period from April 2013 to April 2014 by using spatial and temporal variables. We assume that the daily average concentrations can be divided into two parts: a part dependent on land use variables and a part influenced by dynamic meteorological factors. Due to the small change in land use between 2010 and April 2014, which could be observed from google earth satellite images, the first part can be estimated by annual LUR models developed in 2010. The second part is predicted using two approaches: meteorological factors regression (MFR) modeling and back propagation neural network (BPNN) modeling. The combined LUR + MFR model and LUR + BPNN models were the first attempts at using annual LUR models combined with meteorological conditions to estimate daily variability of NO_2 and PM_{10}.

2. Materials and methods

2.1. NO_2 and PM_{10} measurements

In order to better capture the spatial variation of pollutants concentrations, seventy-four sites for NO_2 and thirty-six sites for PM_{10} were selected based on the location-allocation model described by Kanaroglou et al. (2005). These sites were spread over the study area and represented a range of mild to severe pollution. The distribution of monitoring sites is shown in Fig. 1. Measurements on each site were conducted in four 14-day sampling periods in January, April, July and October 2010, covering each season of the year. Not all measurements were done simultaneously in each season due to lack of a sufficient number of monitoring instruments. To fully capture the effect of pollutants on individuals, each sampler was deployed at a normal breathing height. NO_2 were determined by ethylene diamine dihydrochloride spectrophotometric method and PM10 were determined by gravimetric method, these determinations were conducted in Environmental protection monitoring center of Hunan University. For each site, results from the four measurements were averaged to estimate the annual mean concentrations. Measurements were excluded if the samplers were destroyed or the results from samplers showed obvious inaccuracies. For continuous routine monitoring, NO_2 were collected using chemiluminescence method (EC9841 and TE-42i, Thermo Electron Corp, US). PM_{10} were measured by Tapered Element Oscillating Microbalance (TEOM, RP1400 and 1405, Thermo Electron Corp, US). QA/QC procedures were followed according to Automated Methods for Ambient Air Quality Monitoring issued by the Ministry of Environmental Protection of China. The monitoring stations were deployed according to division of function area. There were nine valid routine air monitoring stations in the Changsha urban area, of which five stations were located near major roads and other stations were located in culture, residential and commercial regions. The sampling heights range from 2 to 15 m. The daily average concentrations were collected to model the temporal variability of NO_2 and PM_{10}.

Table 1 summarizes statistics of NO_2 and PM_{10} measurements in 2010 and from April 2013 to April 2014. Annual mean concentrations were 41.7 $\mu g/m^3$ for NO_2 and 78.9 $\mu g/m^3$ for PM_{10} in 2010. Annual average concentrations of NO_2 and PM_{10} in the period from April 2013 to April 2014 were collected from nine continuous monitoring stations. Annual mean concentrations were 44.5 $\mu g/m^3$ for NO_2 and 103.2 $\mu g/m^3$ for PM_{10}. Measurements showed that pollutant concentrations exhibited significant daily variability with a high standard deviation for both NO_2 and PM_{10}.

2.2. Land use variables and buffers

Spatial predictor variables were generated and stored in GIS. Seven categories of potential variables were generated to characterize the traffic conditions and land use types. Traffic-related variables included two categories: expressway (RD1) and major roads (RD2). We used road length to reflect traffic condition due to the unavailability of accurate traffic intensity data for majority of roads. Because land use in the Changsha urban area exhibits complex spatial configurations, we used five major categories: residential land (RES), industrial land (IND), public facilities land (PUB), green space (GRE) and water area (WAT). The Public facilities land includes total area of commercial, recreation, governmental and education lands. Data on road network and land use were provided by the Changsha Municipal Planning Bureau. Circular buffers were created for 0.3, 0.6, 0.9 and 1.2 km radii using ArcGIS. The sketch map of buffers is shown in Fig. 1.

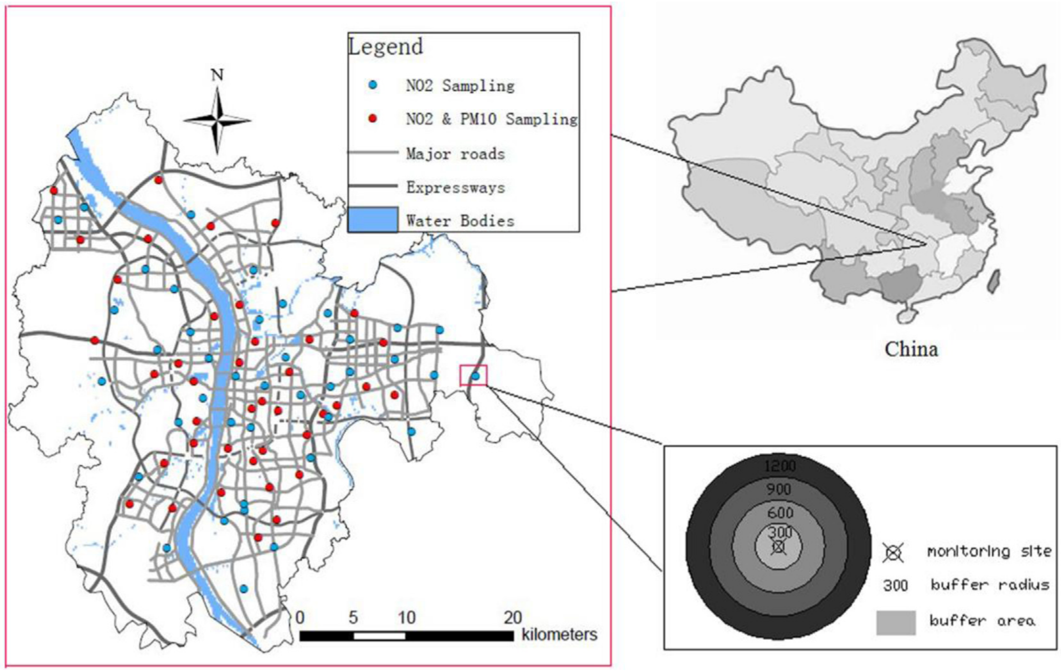

Fig. 1. The distribution of monitoring sites and buffers.

Table 1
Descriptive statistics of NO_2 and PM_{10} measurements.

Pollutants	Measurements (spatial)[a]				Measurements (temporal)[b]			
	N[c]	Mean	Range	SD[e]	N[d]	Mean	Range	SD
NO_2(μg/m^3)	74	41.7	25.5–59.6	8.4	365	44.5	21.7–98.1	16.9
PM_{10}(μg/m^3)	36	78.9	57.3–89.6	9.8	365	103.2	28.6–273.9	31.7

[a] Measurements were averaged over 4-week monitoring periods for each of the 74 sites for NO_2 and the 36 sites for PM_{10}.
[b] Measurements were averaged among nine monitoring stations for each day of the annual period.
[c] Number of monitoring sites.
[d] Number of monitoring periods.
[e] Standard deviation.

2.3. Meteorological variables

Changsha, the capital city of Hunan province, China, is located in a typical subtropical monsoon climate zone. There are four seasons in the Changsha region, typically with a shorter spring and autumn and a longer summer and winter. In this study, nine meteorological variables were generated to characterize the weather conditions, including temperature, relative humidity, air pressure, wind speed, cloud cover, percentage of haze, percentage of mist, percentage of rain and percentage of sun. There is a regular meteorological monitoring station in Changsha urban area, located in north latitude 28.2° and east longitude 113.08°. We collected the daily average values of meteorological data, for some missing values, we used the average of adjacent two days to replace them. The description of data is reported in Table 2. The meteorological data were provided by the Changsha Municipal Meteorological Bureau.

2.4. LUR models

Annual average concentrations of NO_2 and PM_{10} and values of spatial predictor variables were used to develop land use regression models. Both NO_2 and PM_{10} monitoring sites were randomly divided into two groups: a training data set and a test data set. The training data set of 75% of monitoring sites was used to develop the model. The remaining data of 25% of sites was used for model evaluation. Procedures of model development have been described previously (Li et al.,, 2015). Briefly, absolute correlations of each variable with measured pollutants were calculated and the highest-ranking variable in each sub-category was identified. The variables in each sub-category that were correlated ($r \geq 0.6$) with the top-ranked variable were removed from further analyses, and the remaining variables were entered into a stepwise linear regression. In addition, to ensure interpretability of parameters in the final model, an *a priori* criterion was adopted that variables should have intuitive coefficients (i.e., road length should have positive regression coefficients and green space should have negative coefficients). Those variables contradicting the criterion would be removed from the final model. The final equations resulting from the regression are of the form: $Y_a = \beta_0 + \beta_1 X_1 + \beta_2 X_2 + \ldots + \beta_i X_i$. ($Y_a$: pollutant concentrations, β_0: constant intercept, $\beta_{1 \ldots i}$: associated coefficients, $X_{1 \ldots i}$: potential variables). Annual LUR models were evaluated by comparing predicted NO_2 and PM_{10} concentrations with measured concentrations at the retained 25% of monitoring sites. Next, we used the Spatial Analyst feature in ArcGIS to render the regression equations as prediction maps that estimated spatial distributions of NO_2 and PM_{10}.

2.5. MFR models

Meteorological factors regression (MFR) models were developed based on meteorological factors, daily NO_2 and PM_{10} concentrations in the period from April 2013 to April 2014 and annual average concentrations in 2010. Meteorological factors were entered into the models as the independent variables, and the differences between the daily average concentrations in April 2013 to April 2014 and the annual average concentrations of NO_2 and PM_{10} in 2010 were the dependent variable. Both the independent and dependent variables were divided into two parts. The first 20 days of data for each month were used for model development; the remaining part was used for model evaluation. The models were constructed using

Table 2
The description of meteorological data.

Meteorological variables	Units	Range	Mean	Standard deviation (SD)
Temperature	°C	0.1–36.3	19.5	9.4
Relative humidity	%	20.6–90.4	64.3	16.0
Air pressure	hpa	995.5–1033.6	1015.4	9.3
Wind speed	m/s	1–4	1.9	0.8
Cloud cover	%	0–100	58.4	37.8
Percentage of haze[a]	%	0–100	10.0	22.3
Percentage of mist	%	0–62.5	5.1	11.3
Percentage of rain	%	0–100	16.3	29.3
Percentage of sun	%	0–100	68.4	34.8

[a] The ratio of haze monitored in all day.

SPSS software. First, absolute correlations of each independent variable with the dependent variable were calculated. Then, those variables more correlated with dependent variable were retained for further linear regression analysis.

2.6. BPNN models

Back propagation neural network (BPNN) techniques have been widely applied to various types of problems, especially for assessment and prediction. The standard architecture of a BPNN consists of an input layer, at least one hidden layer, and an output layer. The learning algorithm of BPNN applies the fundamental principle of the gradient steepest descent method to minimize the error function (Chen et al., 2010; Xu et al., 2010; Kuo et al., 2013). Philip (1989) suggested that the process of training the BPNN principally includes the following steps: (1) Select the training data set and input the data set to the network; (2) Calculate the output of the network and evaluate the error between the desired output and network output; (3) Adjust the weights within the network based on the gradient steepest descent method that minimizes the error; (4) Repeat steps 1–3 until the error is reduced to a predefined minimal value. Meteorological factors were fed to the neural networks as inputs. The differences of the daily average NO_2 and PM_{10} concentrations in the period between April 2013 and April 2014 and the annual average NO_2 and PM_{10} concentrations in 2010 were entered as output. Before entering into the network, the data were normalized across [0, 1] according to the following equation:

$$R_i = (X_i - X_{min})/(X_{max} - X_{min}) \quad (1)$$

where X_{min} and X_{max} are the minimum and maximum values of the data parameter X_i

In this study, a single hidden layer was used in the BP neural network. The number of hidden nodes was determined by experiment. Data was collected and generated for each day of a year, such that there were 365 samples. Like the MFR model, the first 20 days of data in each month were used for training the BPNN and the remaining samples in each month were retained for testing the accuracy of the trained network. In the neural network program, learning rate and momentum rate were set at 0.5 and 0.7, respectively. The error goal was set at 0.001. The program was run using MATLAB's neural network toolbox.

3. Results

3.1. LUR models

Land use regression models are reported in Table 3. The final NO_2 and PM_{10} models explained 51% and 62% of the spatial variability in measured concentrations. Five to six variables were

Table 3
The LUR and MFR models of NO_2 and PM_{10}.

Model	Pollutants	Equations	N	R^2	R^{2*}
LUR	NO_2 (μg/m³)	$Y_a = 35.7 + \mathbf{RD2.1200} \times 1.16 \times 10^{-3} + \mathbf{RES.600} \times 1.95 \times 10^{-5} + \mathbf{RES.1200} \times 7.59 \times 10^{-6} + \mathbf{PUB.1200} \times 1.20 \times 10^{-5} - \mathbf{GRE.300} \times 3.64 \times 10^{-5}$	74	0.51	0.61
	PM_{10} (μg/m³)	$Y_a = 59.7 + \mathbf{RD1.1200} \times 9.70 \times 10^{-3} + \mathbf{RD2.900} \times 2.50 \times 10^{-3} + \mathbf{RES.1200} \times 3.45 \times 10^{-5} + \mathbf{IND.1200} \times 1.99 \times 10^{-5} + \mathbf{PUB.1200} \times 8.07 \times 10^{-6} - \mathbf{WAT.1200} \times 3.08 \times 10^{-5}$	36	0.62	0.58
MFR	NO_2 (μg/m³)	$Y_b = 27.87 – 0.66 \times$ **Temperature**-4.08 $\times$ **Windspeed** $-$ 13.29 $\times$ **Cloud cover**+21.92 $\times$ **Percentage of Haze**	9	0.43	0.22
	PM_{10} μg/m³)	$Y_b = 58.96 – 1.47 \times$ **Temperature**$-$ 31.47 $\times$ **Cloudcover**+99.57 $\times$ **Percentage of Haze**	9	0.45	0.23

The words in bold denote variables. The first part of the variable name denotes variable type and the second part denotes buffer size, so that RD2.900 = length of major roads within a 900 m buffer. N denotes the number of monitoring sites. R^2 and R^{2*} denotes the determination coefficients for the model and its validation.

entered into the final LUR models, of which the major road, residential land and public facilities land variables explained 46% and 52% of the spatial variability, accounting for the majority of variation for both NO_2 and PM_{10}. To evaluate the performance of the regression models, R^2 for validation and RMSE (Root Mean Square Error) were calculated. For the NO_2 model, R^2 for validation and RMSE were 0.61 and 7.10, respectively. For the PM_{10} model, R^2 for validation and RMSE were 0.58 and 9.00, respectively. Final equations were applied to 20 random validating sites for NO_2 and 10 sites for PM_{10}. Predicted data were plotted against measured data for validation (Fig. 2). The figure shows predicted concentrations were well correlated with measured concentrations for both NO_2 and PM_{10}. Fig. 3 shows regression maps predicting annual concentrations of NO_2 and PM_{10}. The results demonstrate that both NO_2 and PM_{10} concentrations show a discernible spatial distribution. High concentration areas were mainly distributed in the urban center and east region, and low concentration areas were mainly distributed in the north and west regions of Changsha urban area.

3.2. MFR and BPNN models

The final MFR models explained 43% and 45% of the temporal variability of NO_2 and PM_{10} concentrations (Table 3). Temperature, wind speed, cloud cover and percentage of haze were the important meteorological variables in these models. The BPNN models used one hidden layer and 25 hidden neurons. The structure of the network is shown in Fig. 4.

Both MFR and BPNN models were used to model the temporal variability of NO_2 and PM_{10}. To evaluate the performance of MFR and BPNN models, Pearson's r values were calculated between predicted and measured daily average concentrations. In the MFR models, Pearson's r values were 0.43 and 0.47 for NO_2 and PM_{10}, respectively. The correlation coefficients were much higher in BPNN models than in MFR models, with Pearson's r values of 0.82 and 0.92 for NO_2 and PM_{10}, respectively. In addition, predicted concentrations versus measured concentrations from validation are reported in Fig. 5. The results demonstrate that in this scenario the BPNN models were more powerful than the MFR models to explain temporal variability in pollutant concentrations.

4. Discussion

We developed land use regression models in conjunction with meteorological factors regression models and back propagation neural network models for predicting daily average concentrations of NO_2 and PM_{10} in the urban area of Changsha, China. Annual LUR models explained more than 50%–60% of the spatial variability in pollutant concentrations. Compared with model performance in previous studies, with R^2 values ranging from 0.51 to 0.90 for NO_2 and from 0.36 to 0.82 for fine particulate matter (Hoek et al., 2008), our models explained the variability moderately well for both NO_2 and PM_{10} concentrations. The difference in model performance may be attributed to the difference in data quality and land use types. In addition, the lack of small-scale traffic predictors is also

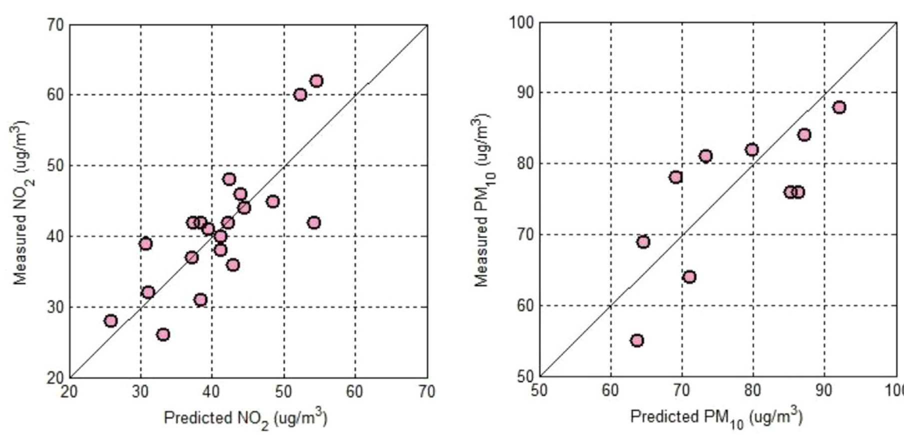

Fig. 2. Predicted versus measured annual average concentrations for NO_2 and PM_{10}.

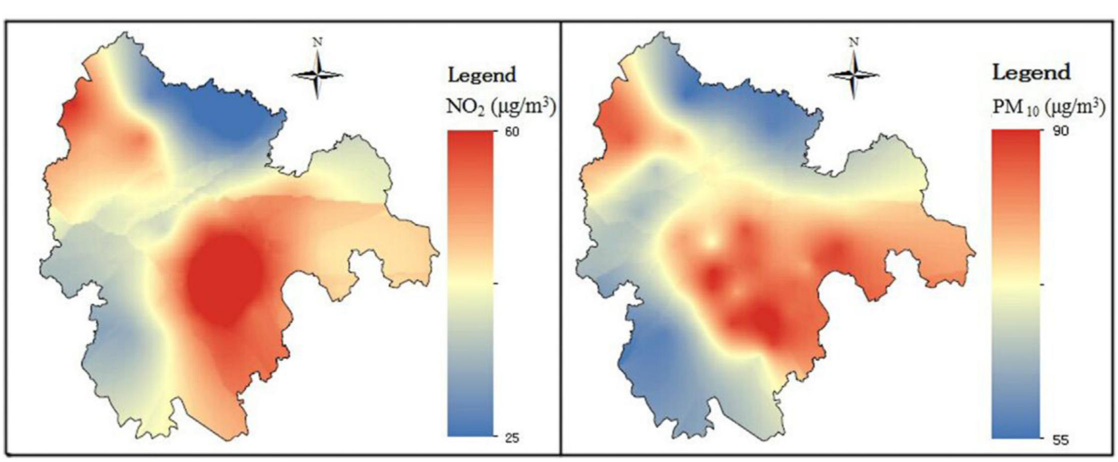

Fig. 3. Estimated annual NO_2 and PM_{10} concentrations from LUR models.

very likely to lead to low R^2 in the model. The spatial variations are indicated by the range and standard deviation of mean concentrations, which were 25.5–59.6 μg/m^3 (SD = 8.4 μg/m^3) for NO_2 and 57.3–89.6 μg/m^3 (SD = 9.8 μg/m^3) for PM_{10} (Table 2). Temporal variations were larger than spatial variations. The range of daily concentrations were 21.7–98.1 μg/m^3 (SD = 16.9 μg/m^3) for NO_2 and 28.6–273.9 μg/m^3 (SD = 31.7 μg/m^3) for PM_{10} (Table 2), indicating the significance of temporal resolution for exposure assessment in epidemiological studies.

Explanatory variables in different LUR models are usually not constant due to city-specific conditions and data availability. Seven land use categories of variables were considered in our model, of which the major road, residential land and public facilities land variables with different buffer sizes explained most of the variability in concentrations, indicating that NO_2 and PM_{10} are closely related to traffic conditions and human activities. Traffic-related predictors were included in almost all LUR models, since vehicle exhaust is a major emission source of NO_2 and PM_{10}. Some studies, including this one, used road length to represent traffic conditions (Beelen et al., 2013; Wang et al., 2013), and some other studies have used vehicle intensity as input data (Ross et al., 2006; Hochadel et al., 2006). Theoretically speaking, vehicle intensity may be better proxy for vehicle exhaust, but complete and accurate traffic intensity data usually are not easily available. Research has suggested that models developed with vehicle density and road length are equally able to explain variability in pollutant concentrations (Henderson et al., 2007). Therefore, we considered road length to be an appropriate variable choice in the absence of vehicle density. Industrial land was not included in the NO_2 model and only explained a small amount of variability in PM_{10} concentrations. This is due to the fact that there are few polluting industrial enterprises in Changsha urban area. In some studies, parameters indicating geographic position were also included such as distance to the coast or a major road (Ross et al., 2006) and longitude and latitude (Jerrett et al., 2007; Henderson et al., 2007), Additionally, population density has proven to be an important variable in some studies (Ross et al., 2007; Ryan et al., 2008). However, these variables were not considered in our models due to the unavailability or high cost of obtaining the data. Despite data limitations hampering our ability to investigate all potential predictors, our regression results still showed a similar degree of explained variability in concentrations compared with other studies, which suggests that a small number of conventional predictors selected according to the specific conditions of a study area are sufficient to build acceptable regression models.

Although many studies indicate that meteorology has a significant influence on the distribution of air pollutants (Arain et al., 2007; Madsen et al., 2007; Wilton et al., 2010), meteorological variables are still not included in most LUR models, possibly due to a lack of an appropriate methodology. In contrast to land use variables showing spatial contrast, meteorological variables show change over time. Therefore, temporal variations in air pollutant concentrations based on meteorological factors may not be stable or easily predictable. This uncertainty may lead to misclassification of exposure assessment in epidemiological studies. For example, if pollutant concentrations are significantly higher or lower in some periods than others, exposure estimates based on data sets in these divergent periods will lead to large misclassification. In this study, MFR and BPNN models were introduced to take weather conditions into consideration to model the temporal variability of NO_2 and PM_{10}. Both models explained temporal variability of pollutants moderately well, indicating that a relationship exists between meteorological conditions and changes in pollutant concentrations. Based on our findings, BPNN models are more powerful than MFR models to explain temporal variability in pollutant concentrations. The results demonstrate that there is a nonlinear relationship between meteorological factors and concentrations of NO_2 and PM_{10}.

For epidemiological studies that examine acute or sub-chronic outcomes, such as a given trimester of pregnancy, due to the temporal variations of pollutant concentrations, the short or

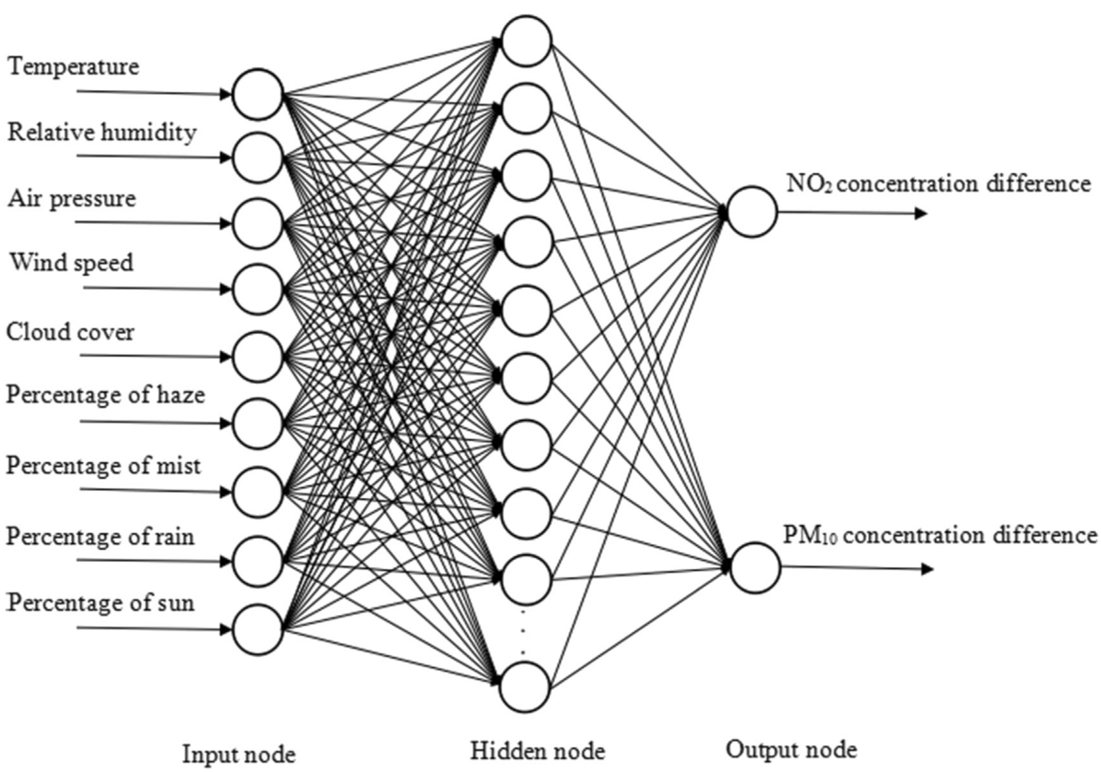

Fig. 4. The structure of the 9-25-2 BP neural network.

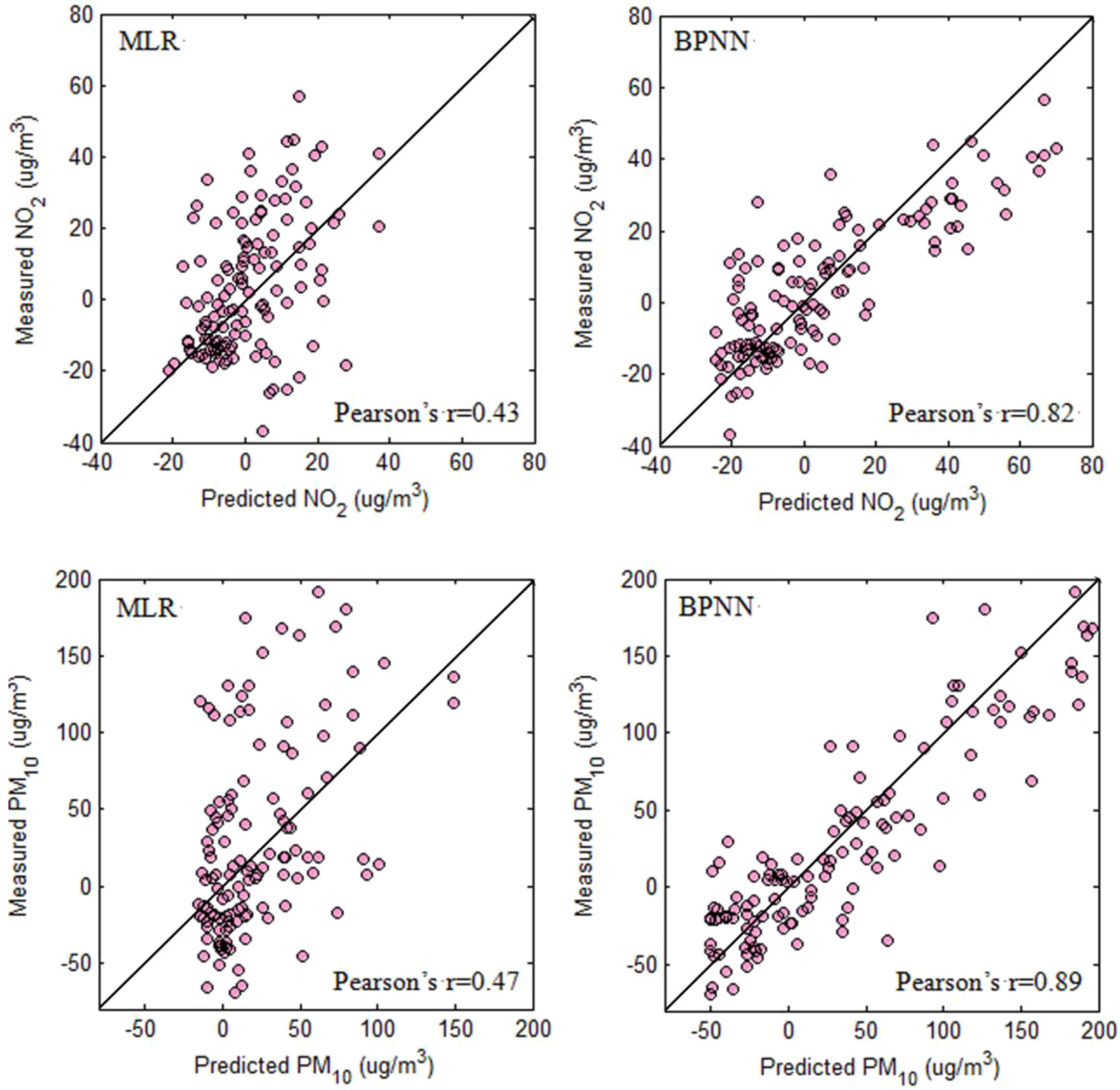

Fig. 5. Predicted versus measured daily and annual average concentrations for NO_2 and PM_{10}.

medium term exposure estimates derived only from annual or seasonal models may lead to large deviations (Wang et al., 2013). Ongoing efforts to estimate quantitative personal exposures have led to different approaches to improve the temporal resolution of LUR models in predicting the variability of pollutants concentrations. The most straightforward way is to calibrate concentrations with observed measurements at a fixed continuous monitoring station (Slama et al., 2007; Mölter et al., 2010). This approach is effective and easy when the study area is located near a fixed monitoring station. However, if the fixed monitoring station is affected by localized pollutant emissions, the predicted concentrations in all other locations would be overestimated and inherently lead to large misclassification of exposure assessment in health studies. Another approach is to build several unique models in different time periods (Gulliver et al., 2011). Each model has different variables and coefficients; as a result, this method requires large amounts of manpower and material resources for data monitoring and collection. The large workload might undermine the cost-effective advantage of using LUR models. In this study, coupling land use variables with meteorological variables to model spatial and temporal variability of NO_2 and PM_{10} was evaluated and showed promising results. By using and refining this method, we can improve our understanding of transfer of pollutants and potential causes of air quality degradation. More importantly, the integrated model is able to predict air quality based on forecasted weather condition and could also predict daily NO_2 and PM_{10} concentrations in those areas with no monitoring site.

The biggest limitations of this study were the weaknesses related to applying the model to a large study area. Our approach presupposes identical temporal trends at every site, although it is expected that meteorological effects will cause non-negligible variation across a large area. Consequently, meteorological variables will show differential influence at each site.

5. Conclusions

In this study, land use regression models and models using meteorological variables were developed and coupled to improve temporal resolution. The integrated models show high spatial and temporal resolution that can provide better exposure assessment in epidemiological studies. Temporally refined LUR models integrated with meteorological variables have the potential to markedly improve the exposure assessment in health studies.

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