JOURNAL OF GEOPHYSICAL RESEARCH, VOL. 114, D20205, doi:10.1029/2008JD011497, 2009

Particulate matter air quality assessment using integrated surface, satellite, and meteorological products: 2. A neural network approach

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Received 20 November 2008; revised 5 August 2009; accepted 17 August 2009; published 30 October 2009.

[1] In recent years, sparse, surface-based air quality monitoring has been improved by using wide-swath, satellite-derived aerosol products. However, satellites are sensitive to the entire aerosol column, not only the aerosol near the surface that impacts human health. In part 1 of this series, we used multiple regression to demonstrate how inclusion of meteorological analyses can help constrain the surface level proportion of the aerosol profile and improve the estimate of surface PM2.5. Here, instead of multiple regression technique, we describe an artificial neural network (ANN) framework that reduces the uncertainty of surface PM estimation from satellite data. We use 3 years of MODIS aerosol optical thickness data at 0.55 μm and meteorological analyses from the rapid update cycle to estimate surface level PM2.5 over the southeast United States (EPA region 4). As compared to regression coefficients obtained through simple correlation (R = 0.60) or multiple regression (R = 0.68) techniques, the ANN derives hourly PM2.5 data that compare with observations with R = 0.74. For estimating daily mean PM2.5, the ANN techniques results in correlation of R = 0.78. Although the degree of improvement varies over different sites and seasons, this study demonstrates the potential for using ANN for operational air quality monitoring.

Citation: Gupta, P., and S. A. Christopher (2009), Particulate matter air quality assessment using integrated surface, satellite, and meteorological products: 2. A neural network approach, *J. Geophys. Res.*, *114*, D20205, doi:10.1029/2008JD011497.

1. Introduction

[2] Satellite remote sensing of aerosols can be used to assess surface level PM2.5 (PM2.5 or $PM_{2.5}$, aerodynamic diameter less than 2.5 μm) mass concentration at high spatial and temporal resolutions [*Al-Saadi et al.*, 2005]. *Fraser et al.* [1984] estimated the columnar sulfate concentration (gm^{-2}) over a few locations on the east coast of the United States using Aerosol Optical Thickness (AOT) retrievals from the Visible Infrared Spin-Scan Radiometer (VISSR) onboard Geostationary Operational Environmental Satellite (GOES). More recently, a study by *Wang and Christopher* [2003] showed that under certain conditions, PM2.5 mass measured at the surface and the 550 nm AOT from the Moderate Resolution Imaging SpectroRadiometer (MODIS) are well correlated (R > 0.7). Although PM2.5 and AOT have different units, they are related to each other through the following equation:

$$AOT = PM_{25}\ H\ f(RH)\,\frac{3Q_{ext,dry}}{4\rho r_{eff}} = PM_{2.5}\ H\ S \qquad (1)$$

where $f(RH)$ is the ratio of ambient and dry extinction coefficients, ρ is aerosol mass density (g m^{-3}), H is the boundary layer height, $Q_{ext,dry}$ is the Mie extinction efficiency, and r_{eff} is the particle effective radius. S is the specific extinction efficiency (m^2 g^{-1}) of the aerosol at ambient relative humidity (RH) [*Koelemeijer et al.*, 2006]. Therefore, the relationship between AOD and PM2.5 is optimal for cloud free skies with a well-mixed boundary layer height. Since the study by *Wang and Christopher* [2003], several papers have been published that have utilized AOT as a surrogate for estimating PM2.5 mass (Table 1). Table 1 shows that most of these studies [e.g., *Chu et al.*, 2003; *Engel-Cox et al.*, 2006; *Hutchison et al.*, 2005; *Al-Saadi et al.*, 2005] were largely focused on the United States and used MODIS satellite data to estimate surface level PM2.5 mass concentration. The MODIS was designed specifically for aerosol studies with good calibration and state of the art aerosol retrieval algorithms to convert measured radiances to AOT. Other studies [e.g., *Gupta et al.*, 2006, 2007; *van Donkelaar et al.*, 2006; *Koelemeijer et al.*, 2006; *Kacenelenbogen et al.*, 2006; *Kumar et al.*, 2007, 2008] also analyzed the MODIS AOT over other parts of the world such as India, Hong Kong, Australia, and Europe. MISR on board Terra also provides reliable AOT retrievals [*Diner et al.*, 2001] and this data has been also used to characterize PM2.5 mass over the selected regions in the United States [e.g., *Liu et al.*, 2004].

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0148-0227/09/2008JD011497

Table 1. A Comprehensive Literature Survey on Satellite Remote Sensing of Particulate Matter Air Quality From Past Half Decade Research

	Reference	Data and Study Area	Key Conclusions/Remarks
1	*Wang and Christopher* [2003]	MODIS, 7 stations, Alabama	Quantitative analysis with space and time collocated hourly PM2.5 and MODIS AOT. Demonstrated the potential of satellite data for PM2.5 air quality monitoring. (R = 0.7)
2	*Chu et al.* [2003]	AERONET, MODIS, PM10, 1 station, Italy	Show relationship between PM10 and AOT. More qualitative discussion on satellite capabilities to detect and monitor aerosols globally. (R = 0.82)
3	*Hutchison* [2003]	MODIS AOT MAPS, MODIS Imagery, GEOS Imagery, PM2.5, Texas	Shows potential of MODIS data in monitoring continental haze over land surface. No correlation analysis
4	*Engel-Cox et al.* [2004]	MODIS, PM2.5 continental United States	First study, which present correlation analysis over the entire United States and discuss difference in relationship over different regions. Qualitative and qualitative analysis over larger area, demonstrated spatial distribution of correlation. Range of R.
5	*Hutchison et al.* [2004]	MODIS AOT maps, Ozone, eastern United States	Used few MODIS AOT maps and discussed the hazy conditions, no correlation analysis, and more emphasis on ozone pollution.
6	*Liu et al.* [2004]	MISR, GEOS-CHEM GOCART, United States	First used MISR data for air quality study and have emphasis on seasonal and annual mean correlation analysis and forecasting. (R = 0.78)
7	*Liu et al.* [2005]	MISR, GEOS-3 Meteorology, United States	Regression model development and forecasting of PM2.5, model generated coarse resolution meteorological fields are used and focused only in eastern United States. 48% explanation of PM2.5 variations.
8	*Al-Saadi et al.* [2005]	MODIS, United States	More descriptive paper on IDEA program, which provides online air quality conditions from MODIS and surface measurements over several locations in the United States
9	*Hutchison et al.* [2005]	MODIS, Texas	Correlation analysis in Texas. Correlation varies from 0.4 to 0.5 and long time averaging can make correlation greater than 0.9
10	*Engel-Cox et al.* [2005]	MODIS, United States	Potential of satellite data for monitoring transport of PM2.5 over state boundaries and event specific analysis.
11	*Gupta et al.* [2006]	MODIS, Meteorology, Global 21 locations	Correlation varies from 0.37 to 0.85 over different part of the world. Cloud fraction, relative humidity and mixing height information can improve relationship significantly. First study covered several global locations.
12	*Engel-Cox et al.* [2006]	MODIS, LIDAR, United States	Weak correlation can be significantly improved by using vertical aerosol information from LIDAR measurements.
13	*van Donkelaar et al.* [2006]	MODIS, MISR, PM2.5, GEOS-CHEM, United States and Global	Intercomparison between MODIS and MISR over several locations in Canada and United States. R = 0.69 (MODIS) and R = 0.58 (MISR). Different approach used to calculate the fine mass concentration.
14	*Koelemeijer et al.* [2006]	MODIS, PM2.5 and PM10, Europe	Mainly focused on Europe. Correlation varies from 0.5 for PM10 to 0.6 for PM2.5. Use of boundary layer height in analysis improved the relationship.
15	*Kacenelenbogen et al.* [2006]	POLDER, France	Intercomparison between POLDER AOT and PM2.5 over 23 sites during April–October 2003. Mean R value is 0.55 with maximum of 0.80.
16	*Liu et al.* [2006]	MODIS, MISR, RUC	Intercomparison between MODIS and MISR in St. Louis area. MISR performed slightly better than MODIS in the region.
17	*Gupta et al.* [2007]	MODIS, Sydney, Australia	Impact of bushfires on local air quality has been studies using both ground and satellite measurements. The quantitative analysis shows up to 10 fold increments in surface level PM2.5 during fires.
18	*Gupta and Christopher* [2008a]	MODIS, Birmingham, Alabama	Provide detailed assessment on satellite remote sensing of air quality. Issues like, MODIS AOT quality flags, cloud contamination, sampling bias, long-term trends, AOT averaging has been discussed using almost 7-year data sets.
19	*Kumar et al.* [2007]	MODIS (5 km), Delhi, India	Three months PM2.5 data from a field campaign were used. Correlation between PM2.5-AOT was 0.52 ± 0.20 ows good agreement between surface and satellite data sets for monitoring air quality.
20	*Kumar et al.* [2008]	MODIS (5 km), Delhi, India	Same as *Kumar et al.*, 2007, plus more analysis on PM10-AOD relationships
21	*Gupta and Christopher* [2008b]	MODIS, southeast United States	Long-term trends in air quality using satellite data could be affected due to sampling bias. Average bias value is about 2 μgm^{-3} on monthly scale for southeast United States.
22	*Apituley et al.* [2008]	AERONET, MODIS, LIDAR, Netherlands	Time varying relationship between AERONET and PM2.5 over single station where R values changes between 0.63 and 0.85. LIDAR data are used to cloud clear AERONET level 1.5 data sets.

Table 1. (continued)

	Reference	Data and Study Area	Key Conclusions/Remarks
23	*Liu et al.* [2008a]	MISR, United States	Describe method of estimating PM2.5 mass and its major constituents using fractional AOD values from different aerosol types in MISR algorithm.
24	*Liu et al.* [2008b]	MISR, United States	Method developed in *Liu et al.*, 2008a is used for case study over EPA STN sites. Attempt to estimate SO_4 and NO_3 is made, which compares well with surface observations.
25	*Martin and Canada* [2008]	Review	Mostly focused on gaseous air quality but also provide some review on particulate matter air quality from satellite observations.
26	*Hutchison et al.* [2008]	MODIS, LIDAR	An attempt is made to improve AOT-PM2.5 relationship by refining MODIS AOT product, optimizing averaging area for MODIS pixels around surface station.
27	*Paciorek et al.* [2008]	GASP, MODIS, MISR, United States	Relationship between AOT derived from geostationary platform and PM2.5. Suggestion to calibrate GASP product for particulate matter applications. R values ranges from 0.41 to 0.51

However, due to its narrow swath width (about 360 km), MISR global coverage is only achieved on a weekly basis and is not suitable for studies that require daily assessment of PM2.5. Recently, AOT data from geostationary satellites at higher temporal resolutions have also been used for estimating near-surface PM2.5 mass [*Paciorek et al.*, 2008].

[3] These studies concluded that the satellite-derived AOT is an important parameter to define air quality over large spatial domains and to track and monitor aerosols sources and transport. Most of these studies are based on correlating AOT and PM2.5 with simple linear regressions [e.g., *Gupta and Christopher*, 2009]. The MODIS derived AOT is a measure of column aerosol loading and cannot be used alone to derive PM2.5 mass concentration, which is an indicator of the mass of the dry PM2.5 near the surface. Meteorological factors such as surface temperature (T_s), relative humidity (RH), wind speed (WS), wind direction (WD), and variations in sunlight due to clouds are important among other parameters, which affect the relationship between the two measures of pollution. Although the AOT-PM2.5 relationships work well in some regions, a major issue is the lack of vertical information as AOT is a columnar quantity whereas the PM2.5 is a surface measurement [*Engel-Cox et al.*, 2006]. Although ground and space-borne lidars are a good solution for obtaining this vertical information, they are not readily available on a daily basis and therefore using meteorological information such as mixing layer heights could be a viable solution [*Gupta and Christopher*, 2009]. Assuming most of the aerosols are in the well-mixed boundary layer this information can be used to refine the AOD-PM2.5 relationships. *Hoff and Christopher* [2009] provide a thorough review of the use of satellite data for estimating surface level PM2.5.

[4] To forecast air quality near the surface, modeling systems are employed that include observations including satellite and ground-based data. These models simulate the emission, transport, diffusion, transformation, and removal of air pollution [e.g., *Mathur et al.*, 2008]. However, daily air quality forecasting based on PM2.5 mass using numerical models is not mature and remains under development [e.g., *Kondragunta et al.*, 2008]. Uncertainties exist because the sources of pollutants are not well defined and also due to gaps in our knowledge of physical, dynamical and chemical processes in the atmosphere. New approaches and systematic modeling are needed to estimate the air quality in the United States and around the world, especially in areas that experience poor air quality that have limited or no ground measurements.

[5] Satellite data have been used to form regression models [*Gupta and Christopher*, 2009], but regression equations tend to predict the mean better than the episodic events and they will likely under predict the high concentrations and over predict the low concentrations [*Dye et al.*, 1998; *Hubbard and Cobourn*, 1998; *Ryan*, 1995]. To explore these issues, an artificial neural network (ANN) based model was developed using satellite, ground and meteorological data sets to assess PM2.5 air quality. In this paper we compare the results of the ANN method with the two-variate (TVM) and multivariate methods (MVM). The TVM and MVM are most commonly methods used to estimate PM2.5 using satellite AOT's (see Table 1). The TVM and

MVM are fully described by *Gupta and Christopher* [2009].

2. Artificial Neural Networks

[6] The complexity of a problem and its understanding decides what type of modeling system is required. A full physically based numerical model would be most suitable for forecasting PM2.5 mass if we have the required data sets (especially emission inventories) and a good understanding of PM2.5 formation and removal processes. However, given the complexity of the problem, a statistical approach is a good compromise [*Gardner and Dorling*, 1998]. ANN is an information processing archetype that was inspired by the way biological nervous systems, such as the brain, process information [*Aleksander and Morton*, 1995]. In other words, ANN is a set of computer algorithms designed to simulate biological neural network in terms of learning and pattern recognition. ANN has been used in many scientific disciplines to identify patterns and extract trends in imprecise and complicated nonlinear data.

[7] ANN has been used for studying various Earth science problems including cloud detection for Polar Regions where traditional methods that employ thresholding algorithms often fail [*Lee et al.*, 1990]. ANN has also been used to specifically investigate forecasting pollution levels in urban areas [*Comrie*, 1997; *Gardner and Dorling*, 1998; *Ruiz-Suarez et al.*, 1995; *Perez et al.*, 2000; *Dorling et al.*, 2003; *Ordieres et al.*, 2005; *Jiang et al.*, 2004; *Perez and Reyes*, 2006; *Chattopadhyay and Bandyopadhyay*, 2007]. A study by *Gardner and Dorling* [1998] in London used ANN to successfully demonstrate the prediction of NO_x and NO_2 by providing meteorological inputs and traffic flow data. Daily mean PM2.5 mass was forecasted in El Paso (United States) and Ciudad Juarez (Mexico) using three different types of neural networks, and two types of models (persistence and linear regression) and results indicated that ANN outperformed the classical statistical models [*Ordieres et al.*, 2005]. *Perez et al.* [2000] compared ANN and classical models to forecast hourly PM2.5 mass concentration using prior day observations in Santiago, Chile. Results from this study again confirm that ANN performed extremely well. These studies concluded that ANN-based modeling systems perform more efficiently when compared to linear regression models for particulate matter air pollution monitoring and forecasting [*Perez and Reyes*, 2006].

[8] In this paper, several NN-based models (or networks) have been developed to estimate surface level PM2.5 using satellite and meteorological fields for various seasons and regions over the southeastern United States. In our previous study, we have used two-variate (TVM) and multivariate methods (MVM) to estimate the surface level PM2.5 mass concentration [*Gupta and Christopher*, 2009]. The MVM results show an improvement in PM2.5 estimation accuracy (13 and 17% for hourly and daily average, respectively) due to the use of meteorology. To our knowledge this is among the first efforts that utilize aerosol information from satellite coupled with surface meteorology to train an ANN system to estimate hourly and 24 h PM2.5 mass concentrations.

[9] In the past, many studies have shown the application of multilayer perceptron (MLP) type of neural network to model air quality and other atmospheric problems. Multilayer perceptron is a feed-forward neural network architecture, which shows the directionality of information processing inside the network. A neural network has the capability of learning a particular skill (such as pattern reorganization and classification) rather than memorizing the training data. For example, training data in air quality applications is usually composed of local meteorological conditions, climatological value of pollution level, along with space and time information that determines the pollution. A trained NN system behaves in a more generalized manner. This is one of the important advantages over linear regression models [*Ordieres et al.*, 2005]. The MLP does not make any assumptions about the data distributions, which is common in other type of statistical methods [*Schalkoff*, 1992] that employ regression methods.

[10] Common neural networks architecture have three layers of neurons: input layer, hidden layer and output layer. Each one of these layers can have one or more than one nodes or neurons. Figure 1 provides a schematic of such a network used in the current study with eight nodes (i.e., input parameters) in input layer and two nodes (i.e., PM2.5 for hourly and 24 h average) in the output layer. The input layer consists of eight nodes; namely, latitude, longitude, month, AOT, wind speed (WS), relative humidity (RH), Height of the Planetary Boundary Layer (HPBL), and surface temperature (TMP). The input layers are connected to the hidden and output layers by a linear combination of functions. Layers between the input layer and output layer are usually called hidden layers and work toward minimizing the error by modifying weights through the training process. Nodes or neurons of a neural network are connected by output signal and weights, which are modified by a simple nonlinear transfer or activation function [*Gardner and Dorling*, 1998]. The MLP needs to be trained using training data sets to predict/estimate outputs. The most common training algorithm is back propagation [*Rumelhart et al.*, 1986; *Hertz et al.*, 1991] where input data are repeatedly sent to a neural network. During each pass of data, the neural network calculates the output (in this case PM2.5 mass concentration), which is compared with the desired output (actual PM2.5 measurement) and an error is estimated. This error is then sent back to the network, which forces the network to adjust its weight such that the error decreases with each iteration until the desired outcome is achieved. Training is therefore the process of finding optimal value of weights for minimizing error functions. Once the optimal weights are obtained, the process of training is completed and the network is ready to estimate/forecast with a new input vector. A step by step description of the training process can be found in the work of *Gardner and Dorling* [1998], and further theoretical details can be found in the work of *Bishop* [1995].

3. Data Sets and Network Training

[11] The data sets are discussed in detailed in part 1 of this series [i.e., *Gupta and Christopher*, 2009] and only a brief description is given here for sake of completeness. Three years of hourly PM2.5 mass concentrations (μgm^{-3}) from EPA AirNow network, hourly meteorological fields from rapid update cycle (RUC) reanalysis at 20 km spatial

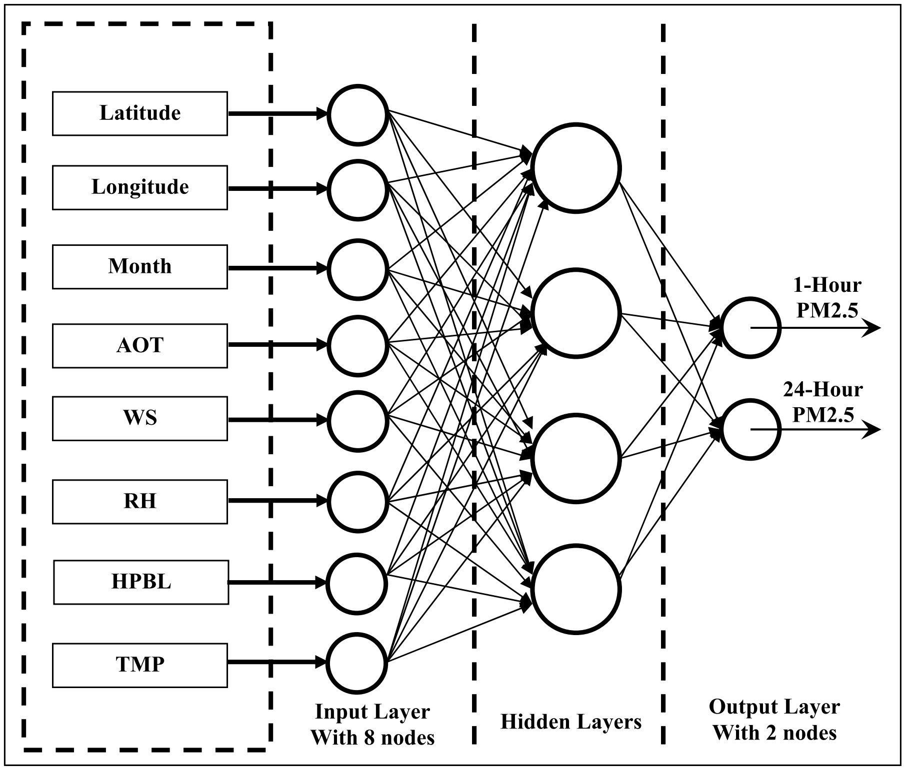

Figure 1. Schematic of a multilayer perceptron neural network used to integrate satellite and meteorological fields to estimate surface level PM2.5 mass concentration.

resolution, and instantaneous retrievals of AOT at 0.55 μm from MODIS-Terra at 10 $\times$ 10 km grid resolution were collected. All three data sets are first collocated in space and time using the methods described by *Gupta and Christopher* [2008a]. Validation exercises [*Remer et al.*, 2005; *Levy et al.*, 2007] have shown that MODIS retrieves AOT over land with a 10–20% uncertainty when compared with ground observations. PM2.5 mass concentration measurements are made using a Tapered-Element Oscillating Microbalance (TEOM) instrument with an accuracy of $\pm$1.5 μgm^{-3} for hourly averages. However, due to the volatilization of ammonium nitrate and organic carbon, the TEOM PM2.5 mass may be underestimated [*Grover et al.*, 2005]. PM2.5 data were collected from 85 ground stations in the southeastern United States. Hourly air temperatures at 2 m (TMP), surface relative humidity (RH), wind speed at 10 m (WS), and HPBL at 20 $\times$ 20 km^2 spatial resolution from the RUC model are used. Intercomparison studies of RUC analysis with METAR (aviation weather reports) provides a RMS difference of 1.5 ms^{-1} and <1.5 K in wind speed and temperature and varies as a function of the season [*Benjamin et al.*, 2004].

[12] This integrated data set, which includes surface, satellite and meteorological information, contains 32,834 samples are used to train, test, and validate the neural network system. Several combinations of neural networks have been trained for different seasons and geographical locations. To construct each of these NN models, the data are divided randomly into three subsets including training (50%), testing (10%), and validation (40%). Training data are used to train the MLP neural networks with multiple hidden layers. Testing data sets are used by neural network to test the performance of learning process after each iteration of the training. Validation data sets were used to perform final validation of estimated PM2.5 values from trained network. Twenty different combinations of NN (models) were trained and the five best were retained for the analysis. The final output and error analysis is produced using an ensemble of these five models. These five models vary in terms of activation functions (see section 2) associated with input layer and hidden layers and on the number of hidden layers used. The number of hidden layers used in the model varies between 2 and 10 where error optimization is performed using sum of square of residual error function. The use of increased number of hidden layers makes the network size large, more difficult to train, and thereby making it slower to operate while increasing the chances of over training. However, at the same time these networks perform better compared to networks with smaller number of hidden layers [*Bishop*, 1995]. Overtraining refers to the reduction in generalization ability of trained network that can occur as networks are trained. In simpler words, overtraining occurs when a network has learned not only the basic relationship associated with input and output data, but also the subtle degree and even the errors specific to the training set. If too much training occurs, the network only memorizes the training set and loses its ability to generalize to new data. The result is a network that performs well on the training set but performs poorly on the validation data and later during actual operation.

[13] Since a large number of training samples ($\sim$15,000) are available for this study, we have allowed the number of hidden layers to vary from 2 to 10. A combination of linear activation functions (see section 2) are used in each of these

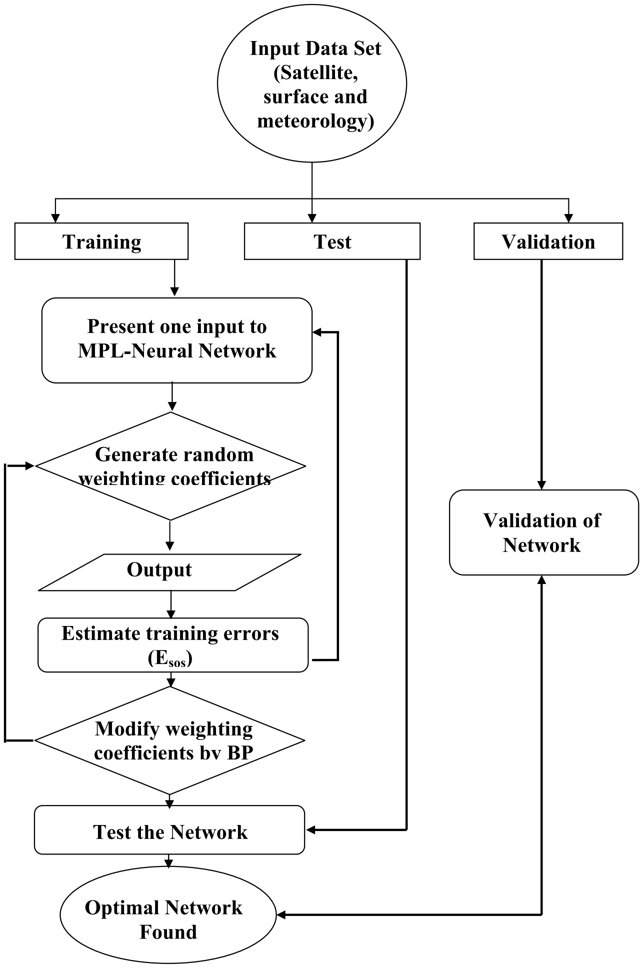

Figure 2. Flowchart describing training process of a multilayer perceptron neural used for surface level particulate matter air quality assessment.

models. The problem of over training is handled using test data sets, which are never used to train the network but rather used to monitor the performance throughout the training process. MLP uses a powerful second-order back propagation training algorithm, which converges quickly to solutions but requires large computer memory. The training of NN is an iterative process where the weighted coefficients associated with each node are modified using new data sets. After every cycle of training, test data sets were used to check the network's output for the desired output (PM2.5 observation) and error function is calculated, which is used to optimize the weights associated with input vector. In this study we use the sum-of-square (SOS) error function, which is given by sum of square of differences between target (desired output) and output (actual output from network) defined over the entire training data set. Equation (2) provides the formulation of SOS:

$$E_{sos} = \sum_{i=1}^{N} (Y_{\mathrm{tar}_i} - Y_{\mathrm{out}_i}) \tag{2}$$

where N is the number of training samples, Y_{tar} is the target value, and Y_{out} is actual output values of the ith sample. Figure 2 presents the flowchart describing entire training process.

[14] Once the network is trained, independent validation data sets that are not seen by the network are used to evaluate the performance of the trained network. The performance of the network is evaluated by calculating absolute percentage errors (APE):

$$\mathrm{APE} = \frac{100 \times \langle |Y_{\mathrm{est}} - Y_{\mathrm{obs}}| \rangle}{\langle Y_{\mathrm{obs}} \rangle} \tag{3}$$

Y_{est} is estimated PM2.5 mass concentration (hourly or 24 h mean) using trained network, and Y_{obs} is observed PM2.5 mass concentration in validation data set. The average difference between observed and estimated value of PM2.5 mass concentration is shown by APE. Absolute percentage error function (equation (3)) is also used to compare the results from neural network model with those obtained using two-variant and multivariant regression models [*Gupta and Christopher*, 2009].

4. Results and Discussion

[15] The results presented here are from the ensemble of five different ANN models that performed the best out of the 20 models trained using training data sets. The current NN has eight nodes (satellite and meteorological fields) in the input layer with varying number of nodes in hidden layer and two nodes (hourly and daily average PM2.5 mass concentration) in the output layer. The number of hidden layers also varies in different networks. The number of nodes in a hidden layer is decided by evaluating the performance of the network by analyzing the errors. Several networks were trained as part of this study, which are described in section 3. Results and evaluation of these networks were performed by statistical measures including linear correlation coefficient (R) and absolute percentage error of estimation (APE) over all the data sets together and as a function of different seasons. Time series of estimated and measured PM2.5 were analyzed over several stations for accuracy assessment as well as for intercomparisons with TVM and MVM model outputs. The PM2.5 mass from both training and validation data sets are shown as scatterplots. Separate analysis is performed for hourly and daily average PM2.5 mass concentration since hourly data sets may not be available in many locations and daily values are used to define air quality standards. We also compare the APE values obtained from NN with those obtained from two-variate models and multivariate models described by *Gupta and Christopher* [2009]. The time series of observed and estimated PM2.5 mass concentration from the three approaches is also provided for selected stations.

4.1. Time Series Examples of Model Outputs

[16] We evaluate the ANN over each station (total 85) by using predicted or estimated hourly and 24 h average PM2.5 mass concentrations. Figures 3a–3c present the time series of observed minus estimated PM2.5 mass (hourly) over three different stations. Figures 3a–3c show the time series of PM2.5 differences of observed and estimated with TVM (red bars, OBS-TVM), MVM (blue open circles, OBS-MVM) and ANN (green squares, OBS-ANN). These stations have

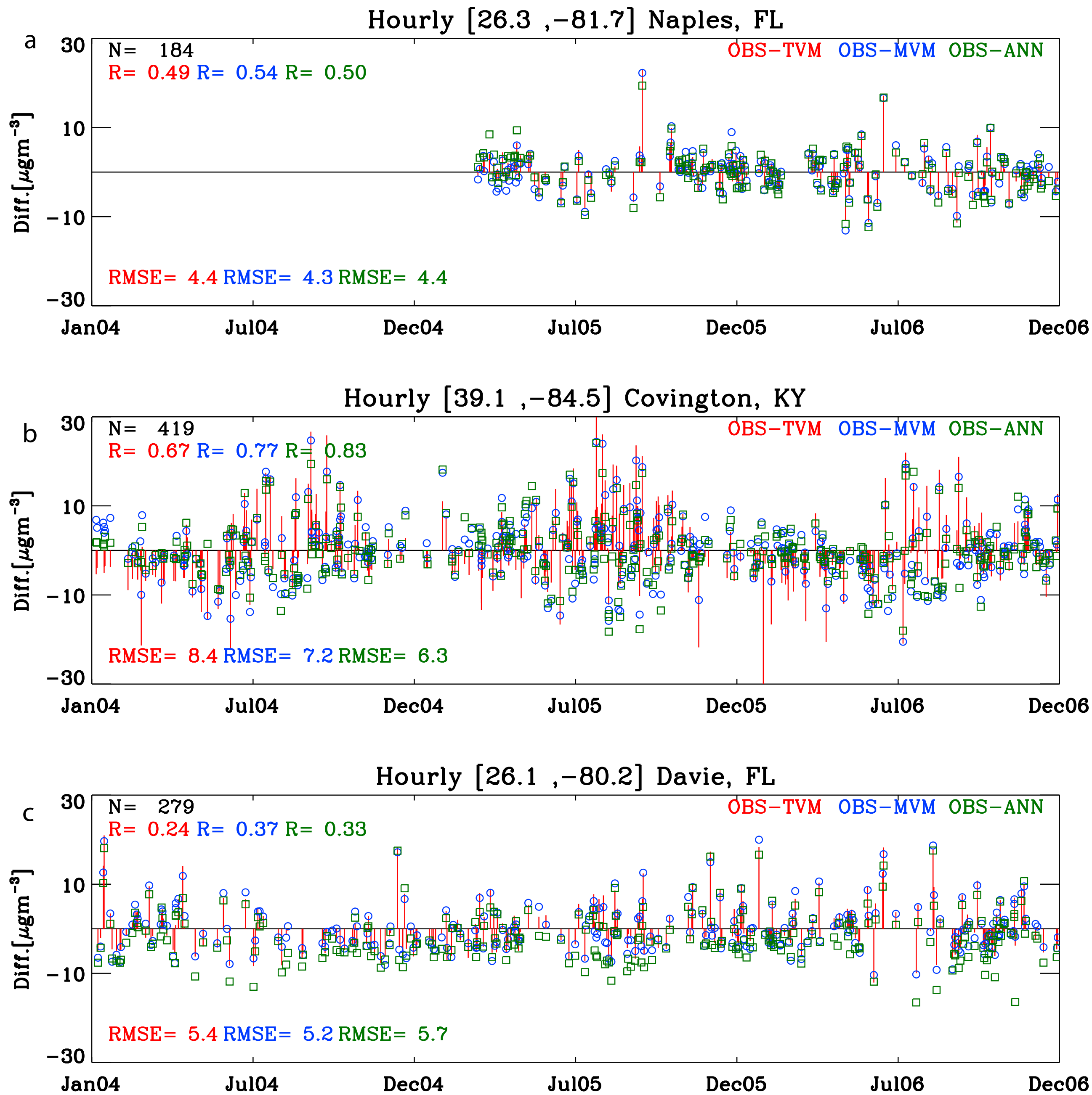

Figure 3. Time series intercomparisons of PM2.5 mass concentration observed from surface station and those estimated using three different statistical models. Difference between observed and estimate from two-variate (red), multivariate (blue), and from artificial neural network (green) are shows in this plot. The three stations are in (a) Naples, Florida, (b) Kentucky-Ohio border area, and (c) Davie, Florida.

been chosen to demonstrate how well (or not) the ANN performs and how it compares with the TVM and MVM.

[17] Station number 1 (Figure 3a) is located in a coastal area [Naples, Florida (26.3 N and 81.7 W; identify as station number 11 in the work of *Gupta and Christopher* [2009]. PM2.5 data over this station is only available during a portion of the study period and remains under good to moderate air quality categories (1–20 μgm^{-3}) with mean value of 8.4 μgm^{-3} with a few high values (>25 μgm^{-3}). Also, this station shows less day to day variability in PM2.5 mass whereas MODIS AOT shows high values in summer and low values during the winter months. The three methods used to estimate PM2.5 produced mean value about the same as observed values, but underestimated them during a few high PM2.5 cases. This station experienced average to low boundary layer height (0.7 km) with highest (1.3 km) values in summer months. The ANN shows no improvement over other TVM and MVM while estimating PM2.5 mass concentration over this site. The correlation coefficient increased from 0.49 for TVM to 0.50 for ANN whereas percentage error of estimation remained the same (39%). MVM, on the other hand, shows slight improvement in the correlation coefficient value (0.54) and a reduction in APE (37%) value. Therefore, none of the three methods per-

formed very well for this station where variability in PM2.5 levels is also observed to be very low. Note that the main input to these models is MODIS AOT (mean AOT = 0.13) where retrieval in coastal areas has larger uncertainties due to the surface heterogeneity [*Remer et al.*, 2005; *Levy et al.*, 2007]. The time series of PM2.5 mass concentrations derived using all three methods does not follow the observed pattern and therefore underestimates some high PM2.5 values while over estimating some very low PM2.5 values. Similar results were observed for other coastal stations. Locations close to coasts often experience transport of aerosols at higher altitude from the remote oceans and land and a possible reason that the columnar AOT is not representative of surface PM2.5 [*Liu et al.*, 2005].

[18] Figure 3b presents a similar time series analysis for a ground station at Covington, Kentucky (39.1°N and 84.5°W, station number 41 in the work of *Gupta and Christopher* [2009]). This station is representative of other stations (>65%) where a good degree of improvement in correlation coefficient and APE value is obtained while using ANN methods as compared to TVM and MVM. Correlation coefficient increased from 0.67 for TVM to 0.77 for MVM to 0.83 for ANN. The APE values reduced to 30% for ANN from 41% and 35% for TVM and MVM, respectively. Figure 3b clearly shows the change in PM2.5 time series behavior when ANN and MVM methods are applied instead of TVM. Root mean square errors (RMSE) over this station are 6.3, 8.4, and 7.2 μgm^{-3} for ANN, TVM, and MVM, respectively. A closer look at the time series indicates that both MVM and ANN methods underestimate the PM2.5 mass during pollution events with high PM2.5 values (PM2.5 > 45 μgm^{-3}). The height of planetary boundary layer over this station reaches as high as 2.3 km with average value of about 0.7 km. The HPBL distribution over this station shows large frequency of HPBL with values greater than 1 km, that improves PM2.5 estimations. Being an urban location (hence dominated by hygroscopic particles), inclusion of relative humidity corrects the AOT values due to enhanced scattering by aerosols particles. This indicates that inclusion of local meteorology and satellite data sets in the model does improve the estimation of PM2.5 mass but the degree of improvement varies with geographic locations, which is a function of aerosol type and uncertainty in aerosol retrievals due to surface characterization.

[19] Figure 3c shows an example of PM2.5 time series for a station where estimation accuracies degraded when ANN was used when compared to MVM method. This is also a coastal station in Davie, Florida and is identified as station number 10 in the work of *Gupta and Christopher* [2009]. Low levels (<15 μgm^{-3}) of PM2.5 mass were observed during the study period with some exception when PM2.5 air quality were in the moderate (>15.4 μgm^{-3}) category. PM2.5 mass concentration estimated using TVM is almost constant with only slight variations. Although, the time series of estimated PM2.5 from MVM and ANN shows some variation, the estimation errors remain high. Application of MVM approach provides some improvement in the estimation of PM2.5 by increasing correlation coefficient from 0.24 to 0.37 and by reducing error (APE) from 49 to 47%. However, the use of ANN approach increased the error to 53%, although there is slight improvement in correlation coefficient (0.33).

4.2. Evaluation of ANN and Comparison With TVM and MVM

[20] Figures 4a–4d present scatterplots between observed and estimated hourly and 24 h mean PM2.5 mass for both training and validation data sets. Also shown are best fit line, the equation of this line including the slope and intercept, linear correlation coefficient R, and number of data points (N). Correlation and intercept values are same in both cases, i.e., when using training and validation data sets for hourly as well as for 24 h averages except for a minor ($\leq$0.01) change in slope values. Therefore, the ANN is producing similar results when using validation data sets which are not seen by the network during the training process. This identical performance of NN on validation data sets shows the proper selection and distribution of training data sets. Our earlier study [i.e., *Gupta and Christopher*, 2008a] has shown that hourly PM2.5 correlated better with AOT as compared to 24 h mean values of PM2.5 mass concentrations. But both MVM approach [*Gupta and Christopher*, 2009] and ANN approach show that 24 h average values are estimated more accurately by these models than hourly values of PM2.5 mass concentration. This could also be due to lower variability in daily mean values compared to large variability in hourly PM2.5 values.

[21] The R values are 0.74 and 0.78 for hourly and daily mean comparisons whereas errors in estimation (APE) are 33 and 24%, respectively. The nearly constant value of intercept ($\sim$6.2) shows the mean bias in the estimated mass concentrations. In Figure 4, the higher values of PM2.5 are underestimated. In some cases overestimation occurs for lower values of PM2.5. MVM method also produced underestimation of PM2.5 mass concentration in the higher range. Earlier speculation for this type of model behavior was the small number (<1% of total samples) of available samples for high (>45 μgm^{-3}) PM2.5 mass concentrations [*Gupta and Christopher*, 2009]. Therefore, current input parameters may not sufficiently represent the association between PM2.5 and independent variables at higher PM2.5 mass concentration. Similar underestimations were also noted by *Liu et al.* [2005] while using MISR aerosol products. For further analysis, the relation between only high PM2.5 and corresponding AOTs is shown in Figure 5. Figure 5 clearly shows that there are many high PM2.5 values between 45 and 60 μgm^{-3} for which AOTs varied from almost 0.0 to 1.4. The small range of PM2.5 mass concentration for a large range of AOT values represents a near constant concentration of surface level PM2.5 with a large variability in the corresponding columnar loading (AOTs). These data points are not associated with the few stations only, but observed over many stations distributed all over the study area. HPBL values during these observations were also distributed over a range from 0.1 to 2 km. This type of AOT-PM2.5 behavior could be due to several reasons including the possibility of multiple layers of aerosols in the atmosphere. In such cases, aerosols that are aloft could also contribute toward the total columnar AOT values. In this case the columnar AOT does not show a good agreement with surface level loading of aerosols. Specific pollution episodes such as biomass burning can produce multiple layers of aerosols aloft up to 5 km in the atmosphere. Mistaking cloud as aerosols in MODIS AOT retrieval algorithm could also lead such AOT-PM2.5

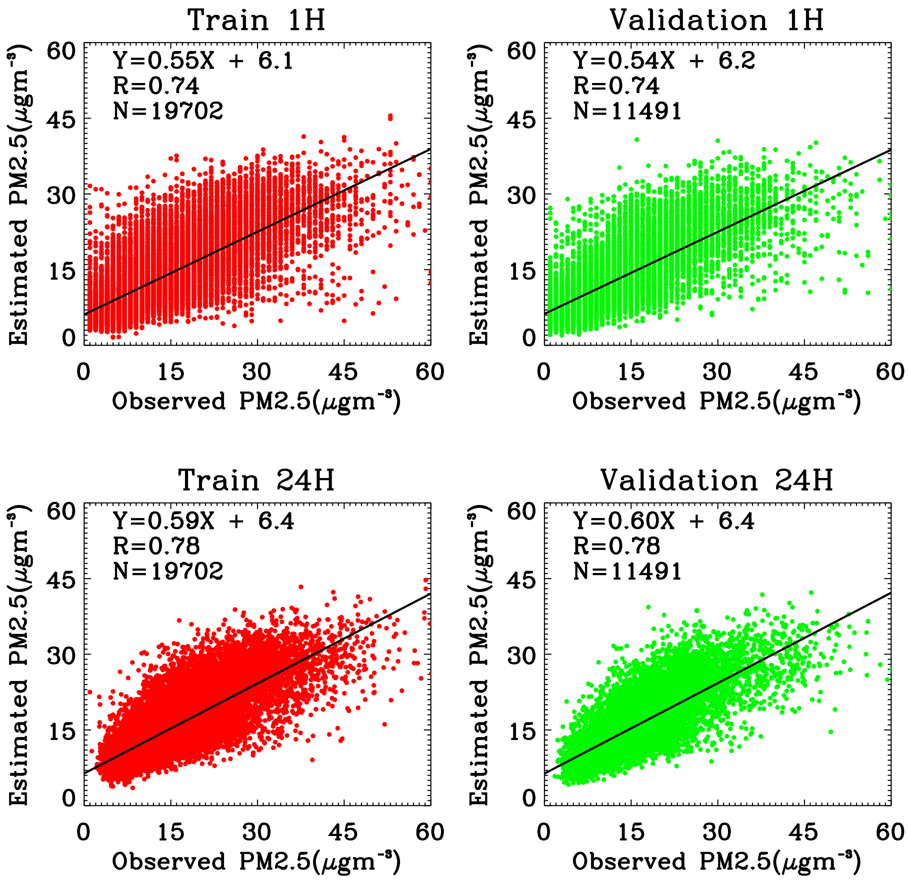

Figure 4. Scatterplot showing performance of trained neural network on all the data sets over a 3-year time period. Separate plots are presented for (top) hourly and (bottom) daily average values of PM2.5. Also, (left) the training and (right) validation (red and green, respectively) data sets are presented.

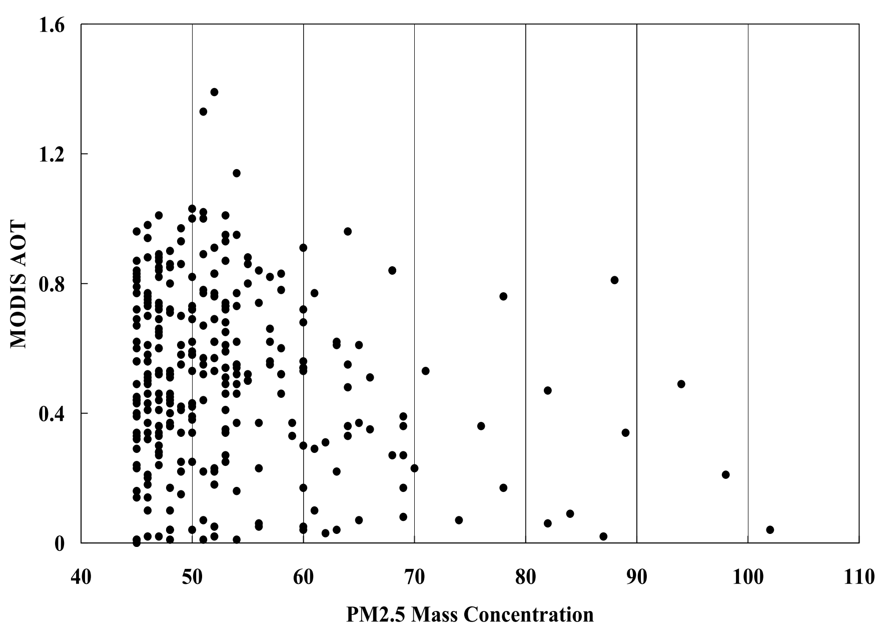

Figure 5. Scatterplot between MODIS AOT and PM2.5 mass concentration (hourly PM2.5 > 45 μgm^{-3}) showing the poor AOT-PM2.5 relationship under high pollution events.

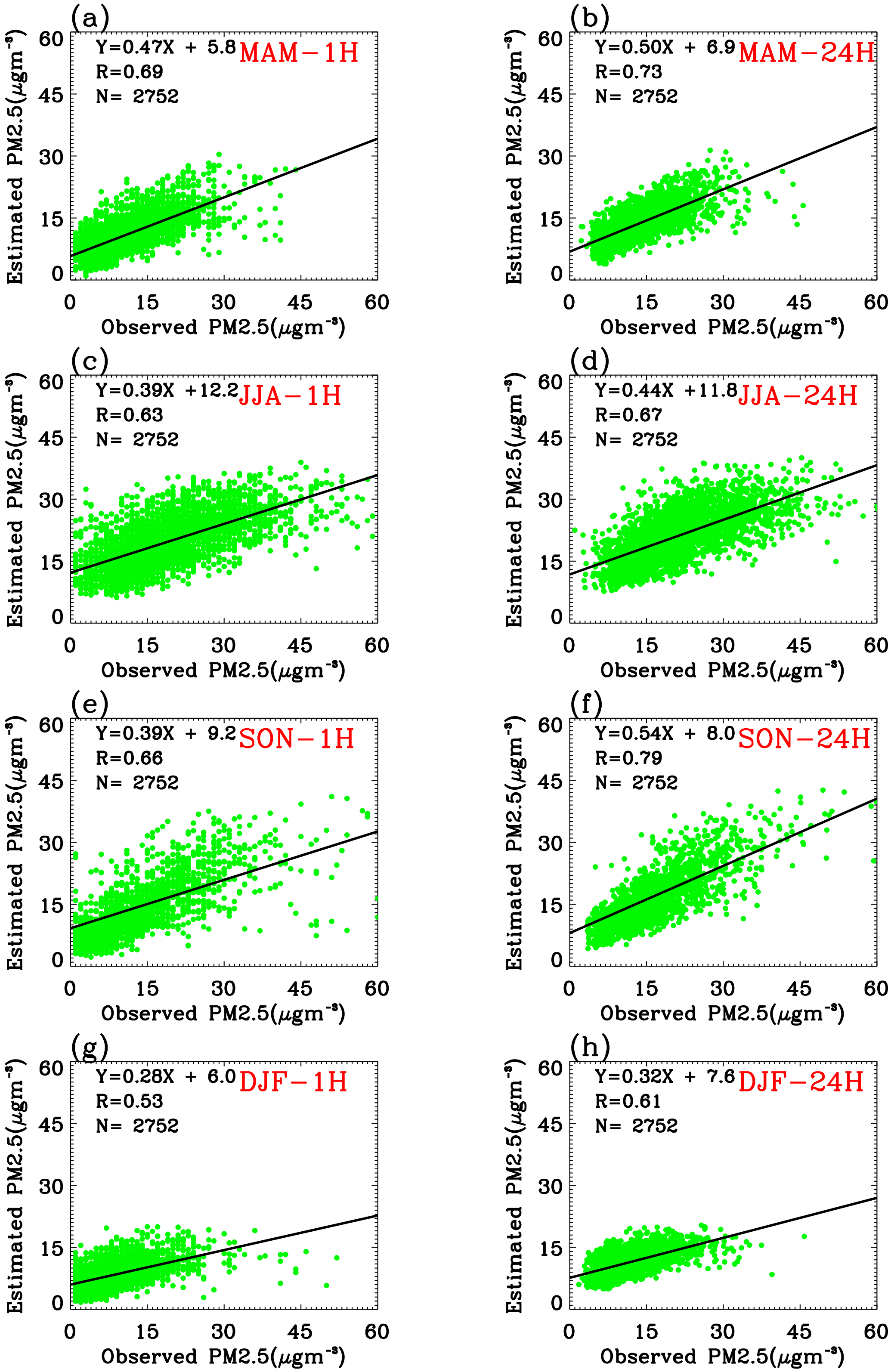

Figure 6. Validation of neural network models trained for each season using training data sets. Scatterplot between observed and estimated PM2.5 mass concentration for hourly and daily average are presented here.

behavior. Another possibility is the change in the particle size distribution due to growth of urban aerosols in the presence of water vapor. Hygroscopic particles such as ammonium sulfate and ammonium nitrate under high relative humidity conditions can grow 2–10 times in size, which increases their light extinction efficiencies [*Wang and Martin*, 2007]. Therefore, high relative humidity conditions also lead to high AOT values while dry mass

Table 2. Absolute Percentage Error Intercomparison of Different Statistical Approach Used to Estimate Surface Level PM2.5 Mass Concentration Using Satellite Remote Sensing Data Sets[a]

		Model Type					
		Two-Variant		Multivariant		Neural Network	
	Data	1 h	24 h	1 h	24 h	1 h	24 h
1	All data	39	29	34 (13)	24 (17)	33 (15)	23 (21)
2	Spring	39	29	34 (13)	24 (17)	32 (18)	22 (24)
3	Summer	34	29	32 (06)	26 (10)	30 (12)	24 (17)
4	Fall	40	30	35 (13)	25 (17)	31 (23)	22 (27)
5	Winter	49	32	44 (10)	28 (13)	41 (16)	26 (19)

[a]The number in parentheses represents percentage improvement over two-variant regression model.

concentration of PM2.5 remains unchanged. Vertical aerosol layer information from spaceborne or ground-based lidars are required to resolve this issue [i.e., *Engel-Cox et al.*, 2006]. Nevertheless, results from this network using all the data sets provide almost 15 and 21% reduction in error (APE) compared to TVM model in the work of *Gupta and Christopher* [2009], whereas improvement in error over MVM method is less than 3 and 4% for hourly and daily average values. Reduction in error over different stations varies within ±10% due to variations in satellite retrieval accuracies and other associated local conditions such as pollution type, emissions sources and transport of pollution.

[22] To understand the seasonal behavior of the data sets using NN, the entire data set is separated into four seasons. Seasons are defined as December to February as winter, March to May as spring, June to August as summer, and September to November as fall. Data for each season is again separated into training, validation and testing data (section 3). Training data set for each season are used to train four different networks exclusively for each season. Figures 6a–6h present the results from validation exercise of seasonal networks developed exclusively for each season. Previous research [*Engel-Cox et al.*, 2004] in this area have shown that over the United States, winter shows weaker relationship between PM2.5 and AOT hence large error associated with PM2.5 estimations. Correlation coefficients for hourly average PM2.5 estimations are highest (0.76) in fall and lowest (0.49) in winter seasons compare to 0.70 and 0.63 in spring and summer. Poor estimation during winter months could be associated with very low HPBL and AOT values, which are subject to larger uncertainties. Under dry stable air during winter when aerosol concentrations are low in the upper atmosphere, high surface concentrations are often confined to very shallow boundary layers, thus limiting the path length for satellite measurements and therefore the sensitivity of the columnar AOT is less. The average height of planetary boundary layer during winter months, was less than 0.5 ± 0.3 km with minimum values as low as couple of tens of meters. MODIS also retrieved very low AOT with an average value of 0.08 ± 0.09 over the entire area during winter months. As discussed in part 1 of this study [*Gupta and Christopher*, 2009], winter month provides non favorable meteorological conditions, such as low temperature and shallow boundary layer, that lead to poor PM2.5 estimations. During summer, the planetary boundary layer is well mixed and is deeper with average value of 0.9 ± 0.5 km [*Gupta and Christopher*, 2009] and the AOTs are well correlated with surface level pollution. R values in case of 24 h average for winter, spring, summer and fall are 0.57, 0.73, 0.67, and 0.82, respectively. Network behavior in different seasons is almost same as MVM but estimation accuracies are improved. The improvement in APE value is greater in case of 24 h average PM2.5 mass concentration estimations (Table 2) since daily data shows low day to day variability compared to hourly measurements.

[23] Tables 2 and 3 provide an intercomparison of the three different methods for estimating PM2.5 mass concentration using satellite data sets. Absolute percentage error of estimation (APE) and linear correlation coefficients (R) are reported in Tables 2 and 3 for each method. The numbers in parenthesis represent the improvement in the statistical parameter over two-variate method (TVM). The greatest improvement in R value for hourly PM2.5 is during winter (133%) whereas lowest improvement is during the fall (23%) season. Similarly, improvement in R for 24 h average PM2.5 is highest in winter (256%) and lowest during fall (32%).

[24] Figure 7 shows the validation of ANN system at a specific station (station number 72). Figure 7 (top) shows time series of hourly average PM2.5 values during MODIS overpass time and Figure 7 (bottom) is for 24 h averages. This is the same location used in part 1 of this study to demonstrate the MVM model performance. This location represents the case where the use of meteorology significantly improved the PM2.5 estimation. Further details on these stations can be found in the work of *Gupta and Christopher* [2009]. Correlation coefficient has changed by 18%, i.e., from a value of 0.73 for TVM to 0.86 for ANN for hourly average values. The percentage error of estimation for this station is 29 and 18% for hourly and 24 h average PM2.5 mass concentration. The observed daily PM2.5 mass shows closer agreement with estimated PM2.5 when compared to hourly PM2.5 values. In general, PM2.5 values over this station are very low (<18 μgm^{-3}) during winter months and it starts increasing in summer (May 2006) and remains high during all summer months. Figure 7 also shows that the agreement between observed and estimated PM2.5 mass are better during summer when compared to

Table 3. Linear Correlation Coefficient Intercomparison of Different Statistical Approach Used to Estimate Surface Level PM2.5 Mass Concentration Using Satellite Remote Sensing Data Sets[a]

		Model Type					
		Two-Variant		Multivariant		Neural Network	
	Data	1 h	24 h	1 h	24 h	1 h	24 h
1	All data	0.60	0.59	0.68 (13)	0.69 (17)	0.74 (23)	0.78 (32)
2	Spring	0.53	0.48	0.64 (21)	0.65 (35)	0.70 (32)	0.73 (52)
3	Summer	0.49	0.49	0.57 (16)	0.61 (24)	0.63 (29)	0.67 (37)
4	Fall	0.62	0.62	0.70 (13)	0.74 (19)	0.76 (23)	0.82 (32)
5	Winter	0.21	0.16	0.42 (100)	0.47 (194)	0.49 (133)	0.57 (256)

[a]The number in bracket parentheses represents percentage improvement over two-variant regression model.

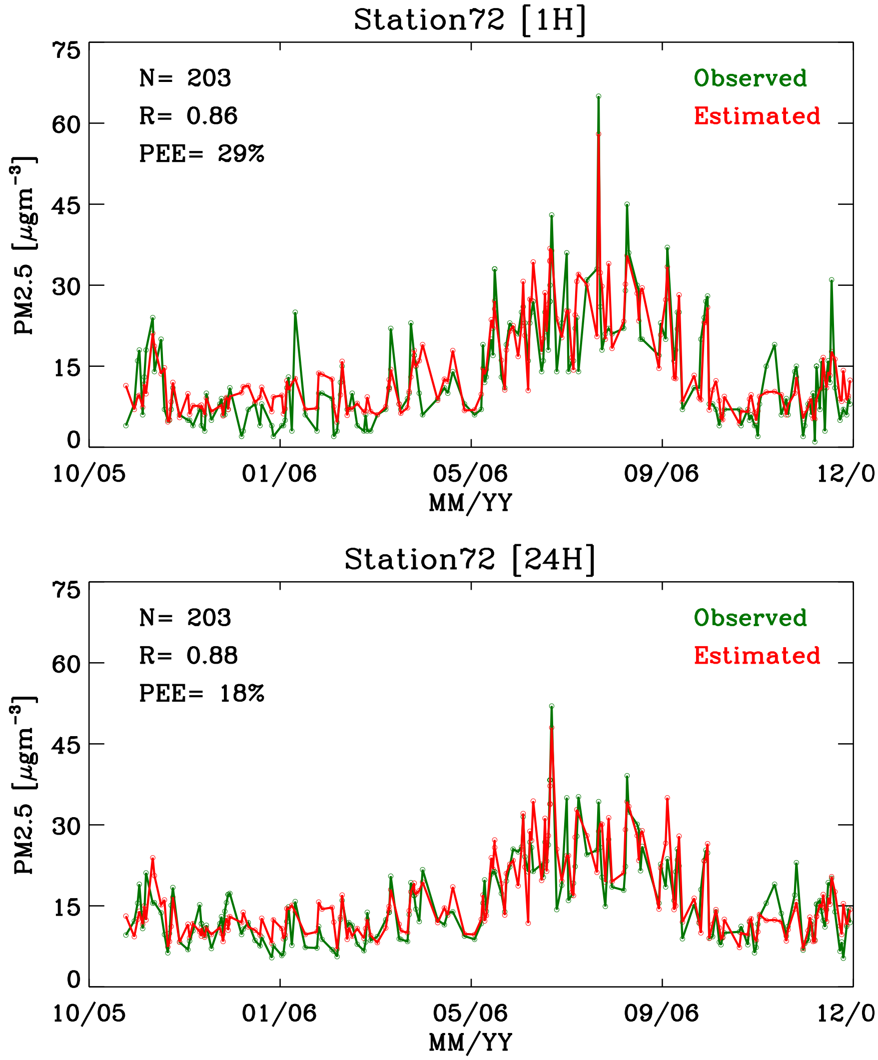

Figure 7. Example of time series validation of model based on neural network for (top) hourly and (bottom) daily average PM2.5 mass concentration estimation over a station in Tennessee.

winter seasons, which is consistent with previous studies [*Gupta and Christopher*, 2008b; *Engel-Cox et al.*, 2004].

5. Summary and Conclusions

[25] Quantitative information on surface level PM2.5 mass concentration is very useful for monitoring and regulating particulate matter air quality. Satellite data are a valuable tool for providing such information over global regions with high temporal and spatial resolutions, especially in the areas where surface measurements are very sporadic or not available [*Hoff and Christopher*, 2009]. The derivation of surface level PM2.5 mass using total columnar AOT value is an ongoing area of research and several challenges remain. In general, AOT-PM2.5 relationships are used to derive PM2.5 mass at the surface. In the current study, we explored the possibility of using an artificial neural network system for estimating PM2.5 mass instead of simple regression equations. Several neural network models have been trained, tested and validated using 3 years of surface, satellite, and meteorological fields in the southeastern United States. This ANN- based model takes satellite derived AOTs and model produced meteorological fields as input and estimates hourly and daily averaged PM2.5 mass. We also compared the performance of ANN model with TVM and MVM models. Results from ANN show significant improvement in APE (15–21%) when compared to the TVM method whereas improvement over MVM is very small (3–4%). The correlation coefficients increased to 0.74 and 0.78 for ANN from 0.60 and 0.59 (which shows an increment of 23 and 32%) for TVM for hourly and daily average PM2.5 mass, respectively. Further analysis shows that improvements in R and APE values vary with seasons as well as geographical locations. The fall months show highest improvement in APE and R values for ANN compared to TVM and MVM models. Absolute percentage errors for fall months show the largest improvements by 23 and 27% on TVM and by 11 and 12% on MVM models for hourly and daily averaged PM2.5 mass concentrations, respectively. ANN models during other three seasons show 6 to 8% improvements in APE values over MVM models. Improvement in correlations during

winter season is almost two to threefolds for ANN over TVM. Overall, the ANN shows more improvement in accuracies for daily averaged PM2.5 mass concentration estimation than hourly averaged values, which is similar to MVM method. Agencies interested in air quality monitoring could benefit from the methods and analysis presented in this paper.

[26] Estimation of daily averaged PM2.5 level is more of interest to environmental agencies for monitoring air quality in the region as their standards are governed by daily averaged values rather than hourly values. The estimation of PM2.5 mass during high pollution level at surface as well as aloft in the atmosphere presents more challenges for all three models. High aerosol loadings in the layer above the boundary layer were found difficult to handle by these statistical models, but inclusion of vertical distribution information of aerosols from new space based lidars such as CALIPSO should improve our understanding. Artificial neural network underestimated high PM2.5 mass concentration, which makes them similar to MVM models. More research is required to model these high pollution events. Further testing of new networks with improved training algorithms, use of new activation functions, more input parameters may produce better results.

[27] **Acknowledgments.** This research is supported by NOAA Air quality projects at UAH. Pawan Gupta was supported by NASA Headquarters under the Earth and Space Science Fellowship (NESSF) grant. MODIS data were obtained from the Level 1 and Atmosphere Archive and Distribution System (LAADS) at Goddard Space Flight Center (GSFC). PM2.5 data were obtained from EPA's Air Quality System (AQS). Special thanks to Dick McNider, Ray Hoff, Arastoo Biazar, and Kirk Fuller for their input.

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