PARTICULATE MATTER AIR QUALITY ASSESSMENT OVER SOUTH EAST UNITED STATES USING SATELLITE AND GROUND MEASUREMENTS

by

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A DISSERTATION

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for the Degree of Doctor of Philosophy
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We, the undersigned members of the Graduate Faculty of The University of Alabama in Huntsville, certify that we have advised and/or supervised the candidate on the work described in this dissertation. We further certify that we have reviewed the dissertation manuscript and approve it in partial fulfillment of the requirements for the degree of Doctor of Philosophy in Atmospheric Science.

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ABSTRACT

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Title Particulate Matter Air Quality Assessment over South East United States using Satellite and Ground Measurements

Fine particles (PM2.5, particles with aerodynamic diameter less than 2.5 µm) can penetrate deep inside the human lungs and recent scientific studies have shown thousands of death occurs each year around the world, prematurely, due to high concentration of particulate matter. Therefore, monitoring and forecasting of surface level particulate matter air quality is very important. Typically air quality measurements are made from the ground stations. In recent years, relationships between satellite derived aerosol optical thickness (AOT) and surface measured PM2.5 mass concentration are formed and used to estimate PM2.5 in the areas where surface measurements are not available. This type of simple linear relationships varies with regions and seasons and do not provide accurate enough estimation of surface level pollution and many studies have shown that AOT alone is not sufficient. Furthermore, AOT represents aerosol loading in the entire column of the atmosphere whereas PM2.5 is measured at the surface; hence the knowledge of vertical distribution of aerosols coupled with meteorology becomes critical in PM2.5 estimations.

In this dissertation I used three years (2004-2006) of coincident hourly PM2.5, MODerate resolution Imaging Spectroradiometer (MODIS) derived AOT, and Rapid Update Cycle (RUC) analyzed meteorological fields to assess PM2.5 air quality in Southeastern United States. I explored the use of two-variate (TVM), multi-variate (MVM) and artificial neural network (ANN) methods for estimating PM2.5 over 85 stations in the region. First, satellite data were analyzed for sampling biases, quality, and impact of clouds. Results show that MODIS-Terra AOT data was available only about 50% of the days in any given month due to cloud cover and unfavorable surface conditions but this produced a sampling bias of less than 2 μgm^{-3}. Results indicate that there is up to three fold improvements in the correlation coefficients (R) while using MVM (that includes meteorology) over different regions and seasons when compared to the TVM and further improvements were noticed when ANN method is applied. The improvement in absolute percentage error of estimation ranges from 5% to 50% over different seasons and regions when compared with TVM models. Overall ANN models performed better than TVM and MVM models. Based on these results, we recommend using metrological variables along with satellite observations for improving particulate matter air quality assessment from satellite observations in the region.

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CHAPTER 1

INTRODUCTION

1.1 Overview

Urban air quality is a critical public health concern in many parts of the globe as urbanization and industrialization have amplified many folds during the last few decades [*NAC*, 2001]. Almost, half of the world's population now lives in urban areas and their number will increase to four billion by the end of this decade [*UNEP*, 1999]. Particulate matter (PM or aerosols) and ozone are two major pollutants affecting the air quality in urban areas of the United States (US) and throughout the world [*EPA*, 2003]. Particulate matter is a complex mixture of solid and liquid particles that vary in size and composition, and remain suspended in the air. Many chemical, physical, and biological components of these PM are identified as being potentially harmful to human respiratory and cardiopulmonary system [*HEI*, 2002]. Particulate matter has many sources from both natural and anthropogenic activities including naturally occurring processes such wind blown dust, dust storms, volcanic eruptions and episodic activities such as forest fires/agricultural burning. Increasing human activities also contribute to combustion from automobiles, industries and emission from power plants [*EPA*, 2003]. Apart from direct

an emission, PM is also produced by other processes such as gas to particle conversion in the atmosphere.

Particulate matter is produced by various sources and therefore their types and spatial variability are different. Pollution in urban areas mainly consist of industrial and automobile emissions but areas closer to the coast also have significant amount of biogenic particulate matter such as pollen, spores, and other sources. The organic particulate matter is mainly a secondary product that forms in the atmosphere by gas to particle conversion. The world's oceans, on the other hand, are the biggest source of salt aerosols in the atmosphere, produced by air bubble bursting at the ocean surface, which depends on near-surface wind speeds. Deserts or semi arid regions of the world produce several tons of dust particles in the atmosphere. Dust particles are in general bigger in size (coarse mode) but there are studies which also report the presence of submicron dust particles in the atmosphere. Dust particles from Sahara desert could be transported to the way to United States and Amazonian and degrades particulate matter air quality [*Chin et al.,* 2007]. Volcanoes are major source of stratospheric aerosols. Volcanoes produce precursor gases (which later convert to aerosols by gas to particle conversion), water insoluble dust and ash in the atmosphere. Biomass burning is another important source of atmospheric aerosols, which produce both particles and gas (which produce secondary particles). Gas to particle conversion mechanism is a very important process which controls the concentration of secondary particles in the earth atmosphere. For example, oxidation of sulfur dioxide into sulfuric acid and its neutralization by ammonia produces sulfate particles. Organic and carbonaceous particles are produced from condensable gaseous matter released into the atmosphere from biomass burning.

1.2 Aerosols and Human Health

Atmospheric aerosols are one of the most important components of the earth-atmosphere system and play important role in climate and weather related processes [*Kaufman et al.,* 2002*; Ramanathan et al.,* 2001]. Particulate matter air pollution has both short-term and long-term effects. Short term impacts include, respiratory infections, irritation to the eyes, nose and throat, headaches, nausea, and allergic reactions. Short-term air pollution can intensify the medical conditions of individuals with asthma and emphysema. For example, in 1952 London experienced one of the worst smog disasters that killed more than four thousand people in few days due to high concentrations of particulate matter in the air [*Scarrow,* 1972]. Long-term effects include lung cancer, heart disease, chronic respiratory disease, and even damage to the brain, nerves, liver, or kidneys. Continual contact to air pollution affects the lungs of growing children and may worsen or complicate medical conditions in the elderly.

Particulate matter with aerodynamic diameters less than 2.5 µm (PM2.5) can cause respiratory and lung diseases and even premature death [*Krewski et al.,* 2000]. The World Health Organization (WHO) estimates that 4.6 million people die each year from causes directly attributable to air pollution [*UNEP,* 1999]. Worldwide, more deaths per year are linked to air pollution than to automobile accidents. Nearly 310,000 Europeans die from air pollution annually [*Leeuwen and Rolaf,* 2002], The Tata Energy Research Institute (TERI) in India estimated 18, 600 premature deaths per year associated with poor air quality in the Delhi region [*TERI,* 2001], increased PM was associated with 2400 deaths per year in Australia with an associated health cost of $17.2 billion [*Morgan et al.* 1998*; Simpson et al.,* 2000] and Sydney experiences around 400 premature mortalities

each year due to increased levels of pollution and asthma is also common in this area [*Barusch,* 1997]. Similar mortality deaths are associated with air pollution in other parts of the world. Direct causes of air pollution related deaths include aggravated asthma, bronchitis, emphysema, lung and heart diseases, and respiratory allergies. A medical study by *Pope III et al.,* [2002] concludes that fine particles and sulfur oxide related pollution are associated with all-cause, lung cancer and cardiopulmonary mortality. The same study also states that an increase of 10 μgm^{-3} in fine particulate can cause approximately a 4%, 6% and 8% increased risk of all-cause, cardiopulmonary, and lung cancer mortality, respectively. Using statistical data collected in twenty big cities, *Samet et al.* [2000] showed that the daily mortality within a metropolitan area is associated with concurrent or lagged daily fluctuations in ambient PM Concentrations. Apart from impact on human health, poor air quality also affects the health of animals and plants. Poor air quality conditions are also associated with damaging buildings and monuments around the world. Indirectly, air pollution significantly affects the economy by increasing medical expenditures and expenditure for preserving the surrounding environment.

1.3 Monitoring Particulate Matter Pollution

Particulate matter air quality is usually measured from ground monitors. In the United States, the Environmental Protection Agency (EPA) monitors air quality by measuring mass concentrations of PM at thousands of ground based monitoring stations across the country. The PM2.5 is measured using a Tapered-Element Oscillating Microbalance (TEOM) instrument with an accuracy of ±1.5 μgm^{-3} for hourly averages. TEOM first collects the particles (<2.5 μm in diameter) on Teflon coated glass fiber filter

surface by passing them through a cyclone inlet, which removes the bigger size particles from the sample of air. The inlet is heated to 50°C prior to particles being deposited onto the filter in order to eliminate the effect of condensation or evaporation of particle water. The filter is attached to a vibrating hollow tapered glass tube. In the mass transducer unit, as the filter progressively load PM2.5 particles, oscillation frequency of glass tube changes proportionally. The change in frequency of oscillation is directly related to the mass of particles on element (filter), which can be measure using computer controlled unit and hence the mass of PM2.5 is obtained in the unit of μgm^{-3} [*Charron et al.,* 2004] under 40-50% relative humidity conditions.

The United States Environmental Agency issues National Ambient Air Quality Standards (NAAQS) for six criteria pollutants namely ozone, particulate matter, carbon monoxide, sulfur dioxide, lead and nitrogen oxides. Standards for particulate matter was first issued in 1971 then revised in 1987 and 1997 by EPA. Recently (September 2006), EPA revised 1997 standards to tighten the criteria. The 2006 standards reduced the 24-hour mean PM2.5 mass concentration standard from $65\mu gm^{-3}$ to $35\mu gm^{-3}$, and retain the current annual PM2.5 standard at $15\mu gm^{-3}$. The EPA reports an Air Quality Index (AQI) based on the ratio between 24-hour averages of the measured dry particulate mass and NAAQS, and it can range from nearly zero in a very clean atmosphere to 500 in very hazy condition. Currently USEPA provides particulate matter air quality forecast over more than 200 cities on daily basis. In recent years, other countries in Europe, Australia, Japan, and China have also started monitoring PM2.5 mass as measure of air quality conditions. However, very few countries have PM2.5 standards. The Australian National Environment Protection (Ambient Air Quality) Measurement (AAQ NEPM) has set

standard values of PM2.5 mass concentration of 25μgm^{-3} and 8μgm^{-3} for daily and annual average respectively (NAAQSTR, 2004). Mexico follows the old USEPA standard of 15μgm^{-3} and 65μgm^{-3} for daily and annual averages. The Canadian Council of Ministers of the Environment (CCME) has set a 30μgm^{-3} daily average PM2.5 value. The World Health Organization (WHO) has recommended air quality guidelines to avoid significant harmful health effects on the human; the daily and annual averages of PM2.5 should not exceed 25 μgm^{-3} and 10 μgm^{-3} respectively [*WHO*, 2006]. However these EPA and other agencies monitoring stations are only point locations and do not have the spatial resolution to map the regional to global distributions of aerosols.

Space-borne measurements are a cost effective way for mapping the spatial distribution of aerosols and their properties due to their reliable, repeated coverage on a near daily basis. Satellite data, for example, aerosol optical thickness (AOT) from MODIS onboard Terra and Aqua have tremendous potential for mapping the global distribution of aerosols and their properties [*Chu et al.*, 2002]. Satellites such as MODIS measure reflected and emitted electromagnetic radiation at top of the atmosphere in several wavelengths covering solar spectrum. Assuming appropriate aerosol models (microphysical and optical properties) radiative transfer calculations are performed to construct look up tables. High resolution (0.5km for MODIS) satellite imager pixels with cloud contamination are first masked [*Martin et al.*, 2002] then averaged over larger area (10 km for MODIS) to improve the signal to noise ratio [*Kaufman et al.*, 1997]. Before averaging over large area, dark surface pixel selection is performed and surface reflectance is estimated using methods described in [*Levy et al.*, 2007]. Reflectance corresponding to each pixel is then matched with reflectance in look table for

corresponding surface conditions and sun-satellite geometry. Aerosol optical thickness is then obtained from look up table, which represents columnar integrated aerosol extinction from bottom to top of the atmosphere. In other words, AOT is measure of aerosol loading in entire column of the atmosphere. However, several outstanding issues remain in using satellite data because most satellite data provide columnar information whereas air pollution near the ground is the most important parameter affecting human health. Several studies have demonstrated the potential of monitoring air quality using high resolution data from space based sensors over regional to global scales. The next section provides summary and key conclusions from these studies.

1.4 Previous Work and Identification of Research Problem

Satellite remote sensing of particulate matter (PM) air quality is a relatively new area of research in the field of atmospheric science. Several research studies have shown the potential of using satellite derived aerosol optical thickness information as a surrogate for air quality conditions. Two salient features of these research are; (1) most studies have used MODIS derived AOT products except few studies by *Liu et al.,* [2004, 2005, 2006], *and Donkelaar et al.,* [2006], which used AOTs from both MISR and MODIS. One of the reasons for the extensive use of MODIS is that it gives much better spatial and temporal coverage as compared to MISR (2) Due to the limited measurements of PM2.5 mass concentration in other part of the world, the area of study in most of the studies has been in some part of the United States except for studies by *Gupta et al.,* [2006], *Koelemeijer et al.,* [2006] and *Donkelaar et al.,* [2006]. The first study by *Wang and Christopher,* [2003] used PM2.5 mass and MODIS AOT data over seven stations in

Alabama and presented very good correlation (>0.7) between these two parameters. This study also concluded that although deriving exact PM2.5 mass from satellite could have larger uncertainties; satellites can provide daily air quality conditions derived PM2.5 with sufficient accuracies. Study *Chu et al.,* [2003] focused on qualitative analysis of MODIS product as an alternative for air pollution in the regions where surface measurements are not available. It also shows the potential of satellite monitoring of transport of air pollution from source to near and far urban areas. Study by *Hutchison et al.,* [2004, 2005] mainly focus on air quality over Texas, and the use of satellite imagery in detecting and tracing the pollution. Study *Engel-Cox, et al.,* [2004] presented a through correlation analysis between MODIS AOT and PM2.5 mass over entire United States. The correlation pattern shows high values in eastern and Midwest portion of the United States whereas correlations are low in western United States. The authors also state that "This variability is likely due to a combination of the differences between ground-based and column average datasets, regression artifacts, variability of terrain, and MODIS cloud mask and aerosol optical depth algorithm" [*Engel-Cox et al.,* 2004]. Their study also concludes that high space and time resolved observations from satellites can provide synoptic information, visualization of the pollution, and validation of ground based air quality data and estimations from models. *Engel-Cox* and co-authors also published other studies in 2004, 2005 and 2006 which further emphasizes the use of satellite derived aerosol products in day to day air quality monitoring and even in policy related decision making. One of these papers [*Engel-Cox, et al.,* 2006] also presented the application of LIDAR derived vertical aerosol profiles to improve PM2.5-AOT relationship. MODIS aerosols and clouds data are now being used in IDEA (Infusing satellite Data into

Environmental Applications) program to monitor air quality over United States. IDEA is a joint effort by NASA, NOAA and EPA to improve air quality assessment, management, and prediction by infusing satellite measurements into analysis for public benefit [*Al-Saadi et al.,* 2005].

The MISR has multi-angle capabilities and provides robust estimates of AOT. However the swath width of the MISR is narrow (360 km) and in the equator and mid-latitude regions global coverage is achieved only a weekly basis. This is a serious limitation for studies that require air quality mapping on daily basis. However, MISR derived aerosol products were first used by [*Liu et al.,* 2004] which shows similar potential for air quality applications. Their study also used GOCART and GEOS-CHEM models derived meteorological fields to examine their relative effects on PM2.5-AOT relationships. *Lie et al* [2005, 2006] also used multi-variant regression equations to evaluate the importance of different meteorological parameters such as planetary boundary layer height, wind speed and direction and relative humidity.

One of the first attempts [*Gupta et al.,* 2006], that compared PM2.5-AOT relationship in different parts of the world such as Europe, Australia, USA, and Asia shows the strengths of satellite derived air quality products at global scales and in the regions where surface PM2.5 measurements are not available. More recently, *Donkelaar et al.,* [2006] published a study, which used GEOS-CHEM derived vertical extinction profiles and basic mass formula to calculate mass of fine particles and compared the results over several locations in USA and Canada. This study also presented global assessment of MODIS and MISR derived PM2.5 mass concentrations.

All these studies mainly concluded that the MODIS and MISR AOTs are important to define air quality over large spatial domains and to track and monitor aerosol sources and transport. These studies are based on correlation and linear and/or multi-variant regression between MODIS AOT, ground based PM2.5 mass and model derived meteorological parameters. The MODIS derived AOT which is measure of column aerosol loading 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 [*Wang & Christopher,* 2003]. Meteorological factors such as surface temperature, relative humidity, wind speed and direction, variations in sunlight due to clouds and seasons are important parameters which affect the relationship between the two parameters. Changes in these processes, which affects the variability in pollution, is primarily governed by the movement of large-scale high and low-pressure systems, the diurnal heating and cooling cycle, and local and regional topography. The vertical profile of aerosol mass extinction, which determines effective scale height and hygroscopic growth factor (a function of rh) are also very important parameters that must be accounted for while deriving relationships between PM2.5 and AOT [*Wang & Christopher,* 2003*; Gupta et al.,* 2006]. Strong winds of 6 ms^{-1} or more can cause dust to become airborne and many factors influence the amount of PM2.5 produced by windblown dust including vegetation cover, soil moisture, soil particle size distribution, surface roughness, and changes in wind direction [*Saxton et al.,* 2000]. Easterly trade winds can transport Saharan dust to the eastern and southeastern US [*Prospero,* 1999] and can increase PM2.5 concentrations at the surface and degrade visibility. Also the western US can be affected by dust transported from Asia [*Falke et al.,* 2001]. Air quality modeling therefore requires a system of models and observations

including satellite and ground-based data that work together to simulate the emission, transport, diffusion, transformation, and removal of air pollution and these models include meteorological models, emission models and air quality models.

My dissertation is built significantly upon these studies by using not only using satellite AOT and ground-based PM2.5 but other ancillary data sets such as meteorological information to monitor and to estimate air quality. A novel neural network based approach is presented to assess particulate matter air quality.

Before satellite and ground-based data sets can be used for air quality studies their quality must be carefully assessed. Satellite data especially must be assessed for cloud contamination and best methods for matching up the ground-monitors must be explored. The first chapter of my dissertation will provide a thorough quantitative analysis of various clouds clearing and match-up criteria. In the second chapter, I will provide an assessment of sampling issues. Since satellite data can only provide aerosol measurements during cloud-free conditions, it is important to assess if this presents a problem. Appendix A will provide quantitative estimates of whether there are sampling biases. Chapter 3 will focus on using meteorological data sets such as winds, temperature, humidity and planetary boundary layer height to improve the PM2.5-AOT relationship. Finally, Chapter 4 will provide a neural-network-based system to assess PM2.5 over the South Eastern United States. Concluding thoughts and future work is also included at the end of the dissertation (chapter 5).

CHAPTER 2

SEVEN YEAR PARTICULATE MATTER AIR QUALITY ASSESSMENT FROM SURFACE AND SATELLITE MEASUREMENTS

2. 1 Introduction

Particulate matter (PM or aerosols) is an important component of air pollution, having both long-term as well as short-term effects on human health such as cardiovascular, lung and skin diseases, which sometimes leads to premature death [*Krewski et al.,* 2000*; Pope et al.,* 2000*; HEI,* 2004*; Pope and Dockery,* 2006]. Particulate matter assessment is of major concern around the world and many environmental protection agencies are working towards continuous monitoring and assessment of air pollution from surface based stations.

Advancement in satellite remote sensing of aerosols over land since the launch of the Moderate Resolution Imaging SpectroRadiometer (MODIS) has provided a new area of research for monitoring global PM air quality [*Gupta et al.,* 2006]. Satellite-derived aerosol optical thickness (AOT) represents integrated atmospheric columnar loading of aerosols and can be used as a surrogate to assess surface particulate matter air quality, especially when surface measurements are not available. Research studies [*Engel Cox et*

al., 2006*; Al-Saadi et al.,* 2005*; Gupta et al.,* 2006, 2007*; Pelletier et al.,* 2007] have shown the potential of using satellite derived AOT values to derive mass concentration of particulate matter with aerodynamic diameter less than 2.5µm (PM2.5). Most of these studies have compared MODIS derived AOT with surface measured PM2.5 mass concentration and linear regression equations were formulated to calculate PM2.5 mass concentration over regions where surface measurements were not available. Some studies have used local meteorological parameters along with satellite-derived products to form multiple regression equations that improve overall relationships [*e.g., Liu et al.,* 2004]. Other studies, have used the Goddard Earth Observing System (GEOS) Chemistry transport model (GEOS-CHEM) derived vertical profiles to obtain boundary layer aerosol optical thickness, which is converted to surface level PM2.5 mass concentration [*Donkelaar et al.,* 2006].

Satellite based studies indicate that AOT data can be used to monitor PM2.5 pollution over global areas on a near daily basis [*Gupta et al.,* 2006*; Chu et al.,* 2003]. Currently, there are several satellites in polar and geostationary orbits that are capable of monitoring aerosols over land from global to regional scales with moderate spatial and temporal resolution. Satellite sensors such as MODIS on Terra and Aqua satellites, Multi-angle Imaging SpectroRadiometer (MISR), POLarization and Directionality of the Earth's Reflectances (POLDER), and Ozone Monitoring Instrument (OMI) are examples of polar orbiting sensors, which provide AOT at 10 to 20 km spatial resolution. Geostationary satellites such as GOES and METEOSAT have also shown the potential of providing aerosol information on much higher temporal resolution [*e.g. Prados et al.,* 2007]. However, most studies use MODIS data, due to its good spatial resolution,

excellent ability to mask clouds, and due to its near-daily global coverage. Although MISR derived AOT values proved to be better than MODIS over land [*Abdou et al.,* 2005], MISR's main limitation is its narrow swath (360 km), allowing for global coverage only every 8 to 9 days thereby limiting its use for air quality studies that require information on daily time scales. Although the MODIS has near daily global coverage, cloud cover and changes in surface properties with season limits the aerosol retrieval on a daily basis.

According to the latest American Lung Association annual air quality report card Birmingham, AL, is ranked as the 4th most polluted city of the United States for particulate pollution. Therefore, we use MODIS (Terra) derived aerosol products along with surface measured PM2.5 mass concentration over North Birmingham (NBHM) to assess particulate matter quality from daily to yearly scales using various methods. We focus on a single air quality station (NBHM) and provide detailed air quality analysis considering different aspects of surface and satellite measurements over a seven year time period, which is unique to this research. Previous studies in this region or elsewhere used data for limited time periods and primarily focused on estimation of relationship between PM2.5 and AOT only. In addition to the PM2.5-AOT relationship, we also examine the differences in surface and satellite observations on monthly and yearly time scales. This provides insight into the effects of aerosol quality flags, on data loss, and we finally provide recommendations on using satellite data to derive particulate matter air quality.

2.2 Data

The data sets used in current study can be categorized into satellite and surface observations. The first is aerosol optical thickness (AOT) data product from Terra-MODIS and the second is daily and 1 hour mean $PM_{2.5}$ mass concentration measured from ground based air quality station using EPA's AirNow network. Both data sets are obtained for a seven-year time period (2000-2006).

2.2.1 MODIS

MODIS Level 2 aerosol data (MOD04, Collection 5) from February 24, 2000 to June 30, 2006 were obtained from the Level 1 and Atmosphere Archive and Distribution System (LAADS) at NASA's Goddard Space Flight Center (GSFC). Each MOD04 granule contains the aerosol properties both over land and ocean retrieved from 5 minutes of MODIS observations using updated collection 5 operational algorithms. Detailed descriptions of these improved aerosol algorithms over land are provided by *Levy et al*. [2007a, 2007b]. The main geophysical validated parameters are spectral aerosol optical thickness (AOT), Angstrom exponent (AE), Cloud fraction (CF), and quality flags related to AOT retrieval, cloud masking, and surface types. Table 2.1 lists the parameters that are used in the current study. We also apply various quality flags associated with cloud masking and AOT retrieval to estimate the difference in mean AOT values with that obtained from standard collocation [*Ichoku et al.,* 2002] and analysis methods [*Wang and Christopher,* 2003] that do not use quality flags. While obtaining coincident MODIS pixels with the PM2.5 measurements, nine different criteria are applied to evaluate AOT

Table 2.1 MODIS aerosol parameters (MOD04, collection 5) used in the current study

No.	Parameter	Description
1	Latitude	Geodetic latitude of each AOT pixel
2	Longitude	Geodetic longitude of each AOT pixel
3	Solar_Zenith	Solar zenith angle corresponding to each pixel
4	Optical_Depth_Land_And_Ocean	Aerosol optical thickness at 0.55 µm for both ocean (best) and land (corrected)
5	Cloud_Fraction_Land	Cloud cover fraction for each pixel determined by spatial technique (Martin et al.,)
6	Cloud_Mask_QA	Cloud mask quality assurance originally derived at 1x1 km that is recomputed at 10X10 km spatial resolution to determine cloudy and clear pixels, land surface type, sun glint, day/night, and snow/ice. Stored as 1 byte SDS
7	Quality_Assurance_Land	Run time quality assurance flag for AOT over land contains product quality flags, retrieval processing flags, and input data resource flags. Stores in 5 byte SDS.

product for PM2.5 air quality applications (Table 2.2). In order to understand these criteria, a brief discussion of MODIS collection 5 land aerosol retrieval is provided.

The MODIS land aerosol algorithm initially retrieves AOT in two wavelengths (0.446 and 0.667 μm) and AOT at 0.55 μm is obtained by interpolating between these two wavelengths. The algorithm also uses the 2.1μm channel along with the vegetation index to obtain surface reflectance [*Levy et al.,* 2007a] and other channels to mask cloudy pixels from each 5 minute MODIS granule. Although the retrieved AOT values are reported at a 10x10 km^2 pixel resolution, the algorithm initially uses radiance measurements at much higher resolution of 0.5x0.5 km^2. All 400 pixels (20x20 pixels), each having resolution of 0.5x0.5 km^2 within the 10x10 km^2 area, are first examined for cloud contamination using a spatial distribution technique [*Martin et al.,* 2002]. The MODIS internal cloud mask [*Ackerman et al.,* 1998] at 1x1 km^2 spatial resolution is also used to identify cirrus. All pixels identified as cloudy are removed and further analysis uses only cloud free pixels to retrieve AOT. Once cloud free pixels are identified, the mean reflectance is calculated from these pixels to retrieve AOT. The measured mean reflectance is matched with lookup tables of calculated reflectance for various aerosol models to obtain aerosol properties for each 10x10 km^2 pixel. *Levy et al.* [2007a] provide further details.

The percentage of cloudy pixels in a 10X10 km^2 grid is computed using 1x1 km^2 cloud mask after applying specific thresholds that are represented in terms of cloud mask quality flags (Cloud_Mask_QA). This Cloud_Mask_QA (CMQA) flag is stored as 8-bit information, which includes percentage cloudy pixels, snow/ice flags and surface type information [*Hubanks et al.,* 2007]. In this study, we have used quality control flags listed

Table 2.2 Criteria used to obtain coincident MODIS –Terra AOT data over PM2.5 locations.

Criteria No.	***Quality Flags***				
	Box size around ground stations	**Surface type**	**SZA (deg)**	**AOT Flag**	**Cloud Flag**
1	±0.25 deg. (~5x5 pixels)	-	-	-	-
2	±0.20 deg. (~4x4 pixels)	-	-	-	-
3	±0.15 deg. (~3x3 pixels)	-	-	-	-
4	±0.15 deg. (~3x3 pixels)	Land	≤ 60	-	-
5	±0.15 deg. (~3x3 pixels)	Land	≤ 60	-	0-90 %
6	±0.15 deg. (~3x3 pixels)	Land	≤ 60	-	0-60 %
7	±0.15 deg. (~3x3 pixels)	Land	≤ 60	-	0-30 %
8	±0.15 deg. (~3x3 pixels)	Land	≤ 60	G & VG	-
9	±0.15 deg. (~3x3 pixels)	Land	≤ 60	G & VG	0-30 %

SZA: Solar Zenith Angle, AOT: Aerosol Optical Thickness

in criterion 4 through 9 (Table 2.2) to determine the importance of cloud contamination on AOT and how it affects AOT calculations for particulate matter air quality research. Aerosol retrieval quality flags (Quality_Assurance_Land or QAL) can also be an important factor on whether or not the retrieved AOT should be matched up with the PM2.5 mass measurements from the ground. This flag contains 40 bits of information, which includes confidence level of retrieval, quality of AOT at both wavelengths (no confidence, marginal, good, or very good), surface reflectance criteria used in retrieval, aerosol types, thin cirrus detection, ozone, water vapor, and snow cover data sources, and other flags associated with the deep blue algorithm [*Hsu et al.,* 2004] In this study, we have used product quality flags from QAL as criteria numbers 8 and 9 (Table 2.2), which shows confidence level of AOT retrieval at two wavelengths.

2.2.2 Particulate Matter Mass (PM2.5)

Particulate matter mass concentration (μgm^{-3}) with aerodynamic diameter less than 2.5 µm (PM2.5) is regularly monitored by United States Environment Protection Agency (USEPA). The particulate matter air quality in the Continental United States is evaluated based on the PM2.5 mass concentration measured using a network of surface stations. Generally, a Tapered Element Oscillating Microbalance (TEOM) instrument is used to measure the mass of PM2.5 particles in units of μgm^{-3}. The EPA and its state partners maintain several air quality monitoring networks in the United States. These networks monitor the mass concentration and speciation (some sites) of gaseous and particulate air pollutants near the surface. PM2.5 data from these networks include 24 h average (daily) concentration data and continuous (hourly) PM2.5 mass concentration measurements. We

use PM2.5 data from the North Birmingham (NBHM, 33.55N and 86.82W) station located in Jefferson County, Alabama, which has a data availability of 85% for any given year. Figure 2.1 shows a detailed view of the stations along with larger view of the EPA region 4.

2.3 Methodology

To obtain coincident PM2.5 and MODIS AOT values at NBHM, nine different criteria are applied (Table 2.2). Mean AOT for each day and each criterion is obtained over almost a seven year time period. The spatial resolution of one MODIS AOT pixel is approximately 10x10 km^2, whereas surface measurements are point values thereby making inter-comparisons difficult. Even if the MODIS pixel was small enough, it does not represent the same viewing conditions due to differences between observation areas, varying path lengths through the atmosphere, and sensor sensitivity to aerosol properties. *Ichoku et al.,* [2002] averaged level 2 MODIS AOT pixels using a 5x5 pixel box over the surface measurement locations and 15-minute observations over one hour to represent a similar air mass as observed by MODIS. This was justified by examining the normal speed of aerosol transport (50 km/hr) using animation of the Total Ozone Mapping Spectrometer (TOMS) imagery over the Atlantic Ocean. This method is used by most satellite aerosol retrieval comparisons with ground measurements [*e.g. Remer et al.,* 2005*; Kahn et al.* 2005*; Abdou et al.,* 2005].

In the current study, three different box sizes (Figure 2.1) centered on NBHM are evaluated to verify the assumptions used in *Ichoku et al.* [*2002*]. Table 2.2 provides all

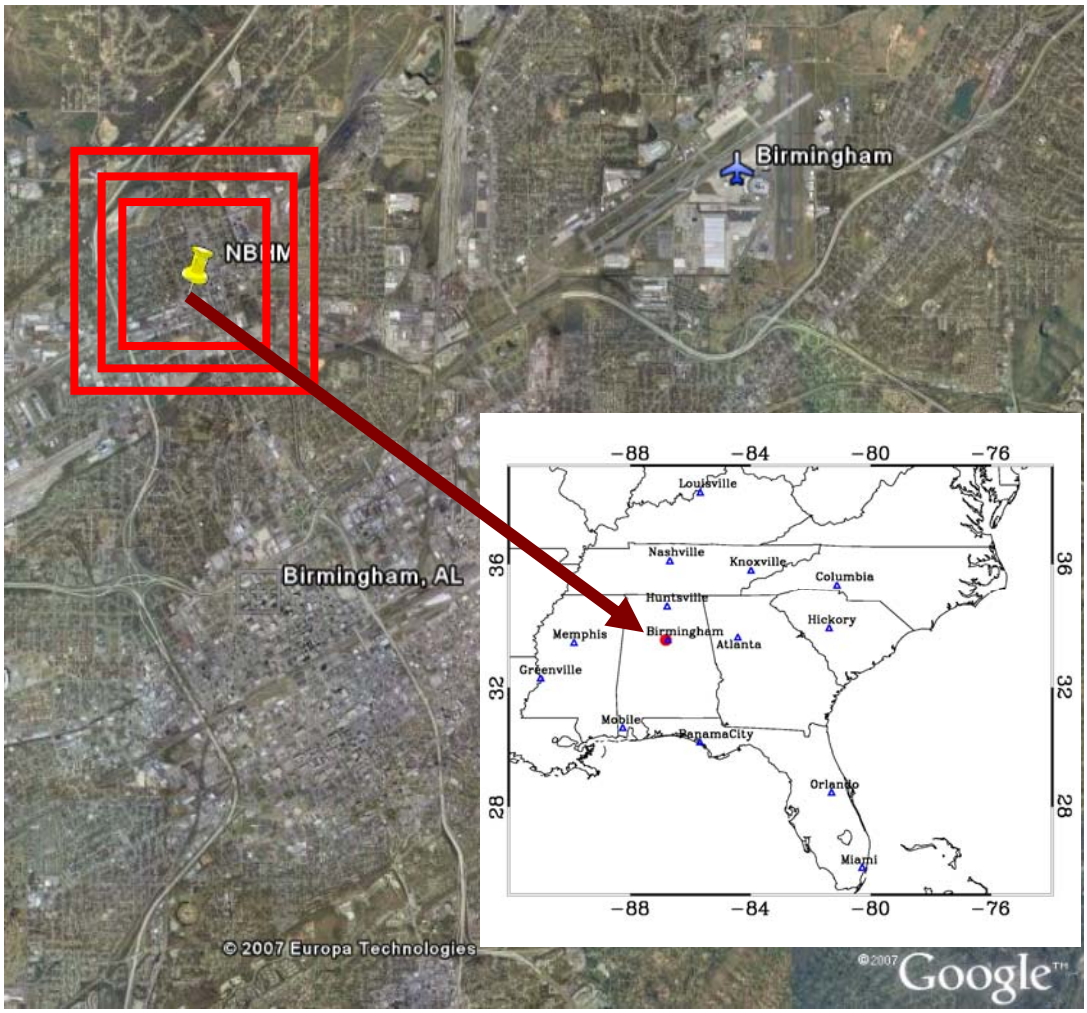

Figure 2.1 Study area with location of PM2.5 ground stations in Birmingham, AL. Also shown is a zoomed satellite image (courtesy Google) of the region with 3 different box sizes used to collocate MODIS observations.

nine criteria used to obtain MODIS AOT. Criteria numbers 1 to 3 represents box sizes of 5x5, 4x4, and 3x3 MODIS pixels around the surface station respectively. It is important to note that in this section, AOT values are obtained for all three conditions without applying any quality flags on MODIS AOT since this is the standard practice in most studies [*e.g. Wang and Christopher,* 2003] that performed such AOT - PM2.5 matches.

Criteria numbers 4 to 9 use the 3x3 pixel box size and apply different quality flags on surface type, solar zenith angle, cloud mask quality and quality of retrieved AOT values. Criteria 4 ensures that all the pixels used in averaging AOT over surface stations are retrieved over land surface with solar zenith angles less than 60°. In some instances there could be inland water pixels, which can create problems in retrieval process or pixels with highly reflecting surfaces might be associated large uncertainty in AOT values [*Hutchinson et al.,* 2005]. Large values of solar zenith angle imply long path length through the atmosphere, increasing the probability of diffuse scattering, which makes estimation of path radiance and atmospheric correction challenging.

Criteria numbers 5 to 7 add the quality flags associated with cloud cover (CMQA) in the 10x10 km^2 MODIS pixels. Cloud mask quality flags are associated with percentage area of 10x10 km^2 pixel covered with clouds based on MODIS 1 km operational cloud mask product [*Ackerman et al.,* 1998]. Criteria 5 allows maximum MODIS pixels for calculating mean value with up to 90% cloud cover, which is reduced to 60% and 30% for criteria 6 and 7 respectively. As the cloud cover criteria is made more stringent, the available number of pixels decreases for calculating mean AOT value, while at the same time the quality of the AOT value increases. However, reducing the number of pixels may also introduce sampling bias into the mean AOT value.

Criteria number 8 corresponds to confidence in AOT retrieval, which is based on number of pixels (with 500 m resolution) used in retrieving AOT at 10 km resolution (QAL). Based on the number of pixels used in calculating mean reflectance to retrieve AOT value, each AOT value is flagged as 'Good' or 'Very Good'. Good and Very Good retrievals for both 0.47 and 0.67 μm channels are considered for this criterion in addition to those used by criteria 4 (Table 2.2). Criteria 9 applies all of the restrictions of criteria number 8 plus, it allows only those pixels for which AOT retrieval is done under less than 30% cloud cover. Criteria 9 uses only the best retrieval situations with all quality flags whereas criteria number 3 represents all retrievals with no quality flags applied to AOT. Collocated AOT values for each of these criteria are tested and correlated with surface measured PM2.5 mass concentration to evaluate the long term change in AOT values as well as PM2.5 mass concentration.

2.4 Results

Results are divided into 4 different sections. Section one discusses the multi-year trends in PM2.5 mass concentration, PM2.5 air quality and MODIS AOT over NBHM. Daily, monthly, seasonal, and annual trends along with frequency distribution of air quality conditions over this site are rigorously examined. The next section explores the availability of MODIS-Terra AOT data over the entire study period and discusses the differences due to satellite sampling in monthly, seasonal, and annual mean PM2.5 mass concentration. This section answers the question of how well satellite sampling (due to cloud cover and other issues) compares with surface measurements (that make continuous measurements regardless of cloud cover) on monthly to yearly time scales.

Section three examines the relationship between MODIS AOT and PM2.5 mass concentration on hourly, daily, monthly, and annual basis. This section also evaluates the stability of AOT-PM2.5 relationships during different seasons and years. The final section, which is unique to the current study, is evaluation of MODIS AOT for PM2.5 research by considering sampling issues as well as quality flags associated with each retrieved AOT pixel. This section also revisits the results of section 1 to 3 under these new criteria for obtaining satellite observations and discusses the differences in AOT and PM2.5 that arise when using stringent quality flags.

2.4.1 Long Term Air Quality Trends over North Birmingham

Surface measured $PM_{2.5}$ mass concentration from NBHM and MODIS AOT from the Terra spacecraft are both continuously available starting in February 2000. Therefore, we have almost seven years of data to analyze daily, monthly, seasonal and annual trends in air quality and aerosol loading over NBHM. Figure 2.2 shows the time series of 24-h mean PM2.5 mass concentration along with monthly and annual mean trends. Daily variations in PM2.5 mass are represented by thin light gray line, which are mainly associated with local meteorological conditions and changes in local emissions of PM2.5 particles. High values could also be associated with transport of pollution from surrounding areas. The background colors denote the air quality conditions as defined by USEPA based on mass concentration of aerosol particles that are smaller than 2.5 µm in aerodynamic diameter. The horizontal red line corresponding to 15 μgm^{-3} and 35μgm^{-3} are the annual and 24 h mean national standards set by USEPA under National Ambient Air Quality Standards (NAAQS), while the previous (prior to prior to December 17,

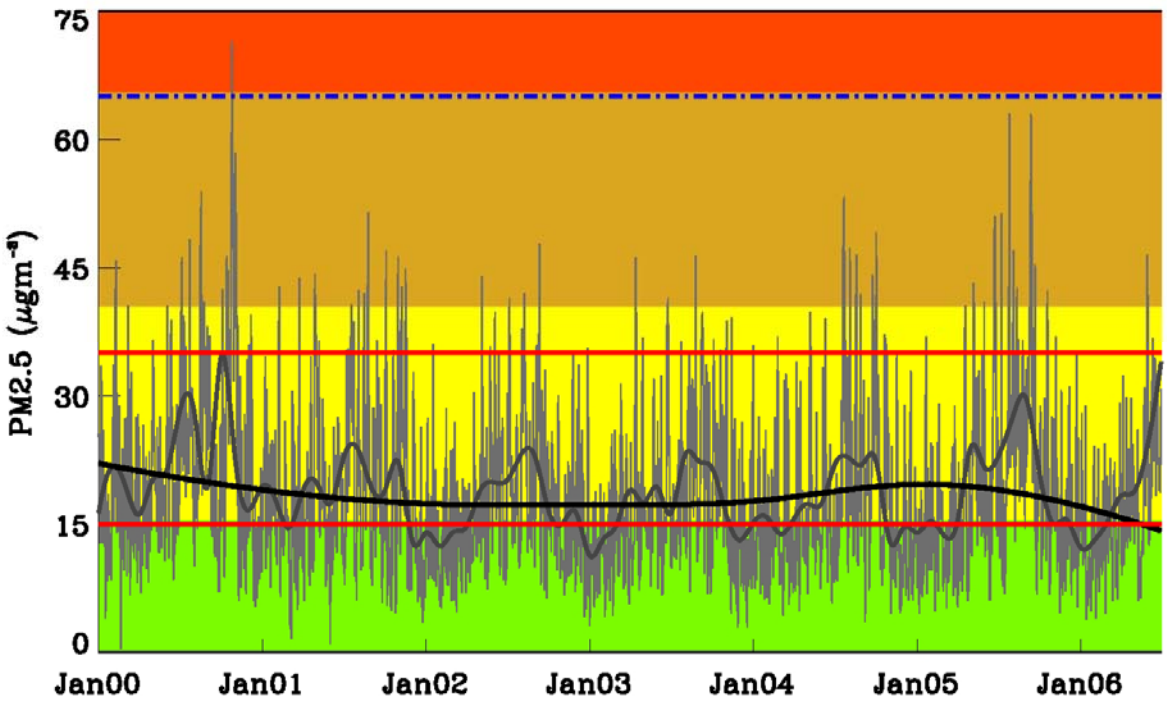

Figure 2.2 Time series analysis of PM2.5 mass concentration over NBHM site starting from January 2000 to June 2006. Background colors show air quality categories. Daily variations are shown in thin light gray line, monthly mean in thick gray line and yearly means are shown in thick black line. Two red lines show new daily and annual national standard for PM2.5 mass whereas dotted blue line shows old annual USEPA standards.

2006) 24 h standard of 65μgm^{-3} is also plotted for comparison purposes (Figure 2.2). Our analysis indicates that NBHM often experiences moderate to unhealthy air quality conditions for sensitive groups, but rarely reaches the unhealthy category even under the new guidelines. The maximum 24 h mean PM2.5 mass concentration of 75.3μgm^{-3} was observed on October 25, 2000 whereas the average of all daily mean values is 18.7±9.7 μgm^{-3}. The thick gray line presents the monthly mean PM2.5 mass concentration calculated using daily mean values. The monthly mean values clearly show a seasonal trend with high values in spring-summer and low values in fall-winter months. High spring-summer values partially correspond to increase in gas to particle conversion in the atmosphere with increasing available solar radiation, enhancing the photo chemical reactions responsible for such particle production. Similar conditions do not occur in the winter months, thereby limiting PM2.5 production. Seasonal changes in local meteorology also affect the PM2.5 production and removal processes. The month-to-month variation is not similar in all the years and varies slightly with peak values around July-August. Monthly mean PM2.5 mass peaks at 34.6μgm^{-3} during October 2000 with a minimum value of 11.3μgm^{-3} observed in January 2003. A weak decreasing trend in annual PM2.5 was noted with the highest and lowest PM2.5 mass concentration of 22.1μgm^{-3} and 17.0μgm^{-3} observed in 2002 and 2006 respectively.

Figure 2.3 presents similar analysis for AOT derived from MODIS using criteria 1 from Table 2.2. Recall that this criterion includes all AOT data without any quality flags. The MODIS AOT shows similar trends with high values during summer and low values during winter months. Seasonal changes in AOT are more prominent compared to seasonal changes in PM2.5 mass concentration due to sensitivity of urban aerosols

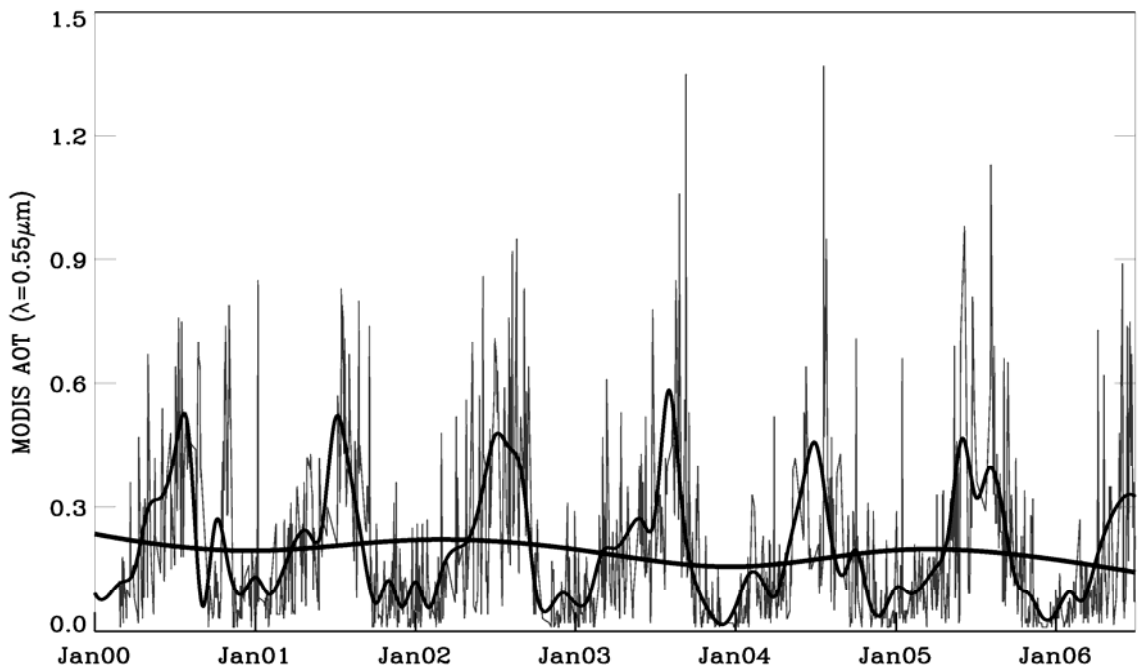

Figure 2.3 Time series analysis of MODIS AOT over NBHM from February 24, 2000 to June 2006. Daily variations are represented by thin light gray line, monthly mean represented by thick gray line and yearly means are shown as thick black line. These results are from criteria number one.

towards relative humidity [*Wang et al.,* 2007]. The absolute difference in PM2.5 mass from winter to summer is not as high (when compared to PM2.5 trends in Figure 2.2) as observed in MODIS AOT. Recall that AOT is an optical property which is function of light scattering from the aerosol particle and it increases in summer time due to particle growth under high humid conditions [*Hess et al.,* 1998]. Therefore, the same aerosol mass can produce large AOT values during summer months compared to winter months. On the other hand, PM2.5 mass does not change significantly due to growth of particles as TEOM make measurement of dry particles under about 40% relative humidity conditions. MODIS AOT also shows overall decreasing trend in annual mean values over study period.

Figure 2.4 presents frequency distribution (days of occurrence) of air quality conditions during each month. Also shown are days of occurrence (%) for each month (secondary y-axis) averaged over all 7 year time period shown as filled circle and interpolated lines. During October 2000, air quality conditions were unhealthy corresponding to the month of highest PM2.5 mass concentration. Frequency of a particular air quality condition shows large variability, but does indicate the presence of a seasonal cycle. Monthly means clearly show that December had the highest frequency of days with good air quality (~65%). The frequency of good air quality begins to decrease in January and generally continues to decrease until August when only 23% of the days have good air quality conditions. Starting September, the air quality condition improves again until December. Moderate air quality conditions occurred 61-68% of the time during the months of July and August with the lowest (35-37%) occurring during December and January. Air quality degrades to unhealthy for sensitive group (USG)

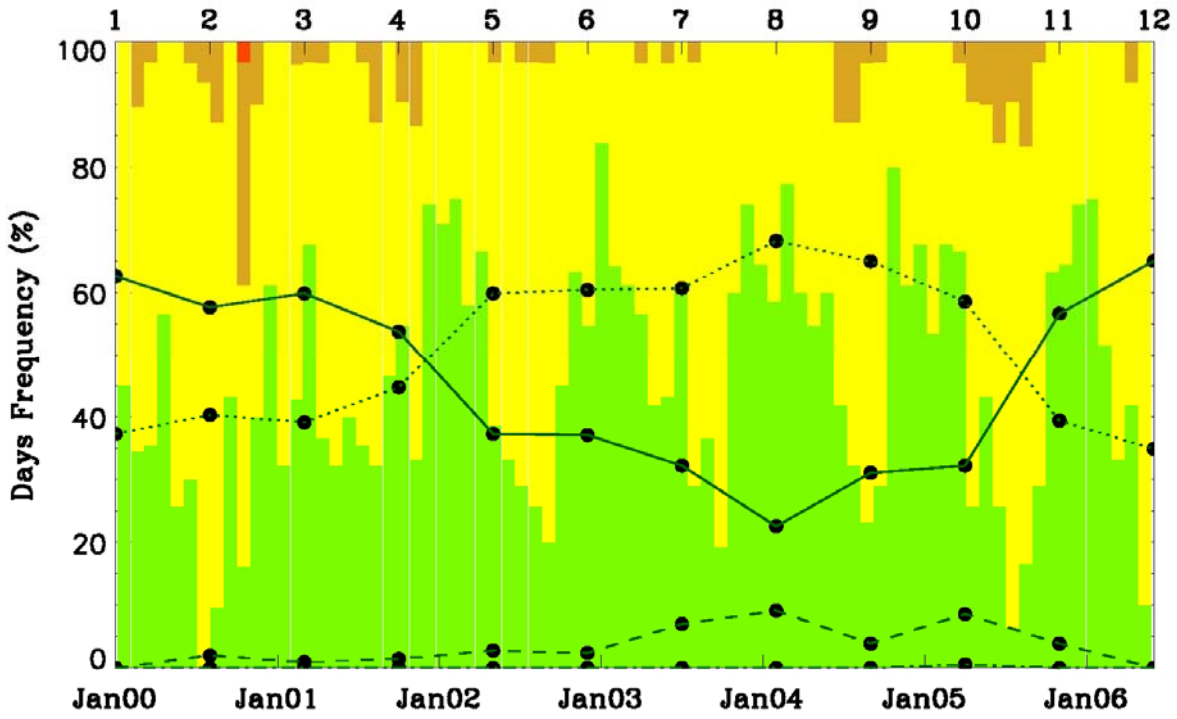

Figure 2.4 Frequency distribution of number of days (%) falling in any particular air quality category during each month over NBHM starting from February 2000 to June 2006. Air quality categories are derived from PM2.5 mass concentration as defined by USEPA and represented by different colors. Green is good air quality, Yellow is moderate, Orange is Unhealthy for sensitive groups, and Red is Unhealthy. Mean frequency for each month over all years is presented as line and open circle for each air quality category and plotted on top axis. Solid line: Good, dotted line: Moderate, dash line: unhealthy for sensitive groups.

category on rare occasions and accounts for 9% days during August, whereas it remains less than 2-3% for other months. Seasonal variations in primary emissions and secondary formation (gas to particle) rates lead to seasonal differences in PM2.5 concentration. Day to day variations in PM2.5 mass concentrations is usually associated with variations in local meteorological conditions. Meteorological conditions that strongly control PM2.5 mass concentration include change in available sunlight due to clouds and season, vertical mixing of air pollution within atmospheric boundary layer, temperature, moisture, long and short range transport by winds and recirculation of air mass by local wind pattern. Very high level of pollution can be observed during strong temperature inversion near surface [*Kukkonen et al.,* 2005] and the magnitude, vertical structure, and temporal evolution of inversions influence the air quality [*Hussein et al.,* 2006].

2.4.2 Availability of Satellite Data for Air Quality

Since the launch of MODIS onboard Terra, almost daily global coverage of AOT retrievals are available and has been used for various climate and air quality research applications. Retrieval of AOT is limited to a cloud-free atmosphere for certain surface conditions, since MODIS cannot retrieve AOT over bright targets such as desert and bright urban centers [*Levy et al.,* 2007a]. Advances in observation capabilities of satellites and improved retrieval techniques are slowly removing these limitations [*Hsu et al.,* 2004].

In this section we explore the following question: 'How well can MODIS Terra AOT data represent monthly and yearly mean PM2.5 mass concentration over NBHM'. To answer this question, we assume that PM2.5 mass can be estimated using MODIS

AOT data. To test this assumption, daily mean PM2.5 measurements are collocated with MODIS AOT data according to criteria 1 as given in Table 2.2. Although the Terra satellite overpasses NBHM site almost every day around 10:30 am local time, AOT retrieval may not be available on a daily basis due to clouds and other limitations (e.g bright surface during certain seasons). Figure 2.5a presents the number of days (%) in each month for which AOT data available over the past seven years. The seven-year mean value is 47 %, but with significant monthly variability. The numbers of available AOT days are mainly associated with cloud free days over the surface station. Terra-MODIS monthly mean daytime cloud cover data reveals that mean fractional cloud cover was 55% in the region. Over the entire study period the maximum number of available days was 90% during October 2000 whereas the minimum was about 7% during August 2005. Number of available days do not show significant seasonal trends but demonstrate high values during fall and winter and low values in spring and summer months. Month to month changes in available days vary from maximum of 57% during October to minimum of 35% in February averaged over all seven year time period. Tracking number of days in each year is important while evaluating long term trends.

Figure 2.5b presents the monthly mean difference in PM2.5 mass concentration between all 24-h mean PM2.5 measurements averaged for each month (ALL) and PM2.5 measurements averaged only when MODIS Terra AOT is available over the measurement site (SAT). The purpose of this analysis is to evaluate how well satellite measurements can represent monthly and year mean values of PM2.5 mass. Negative differences indicate that using PM2.5 data during the time of the satellite overpass overestimates PM2.5 on monthly as well as yearly basis. This indicates that events with

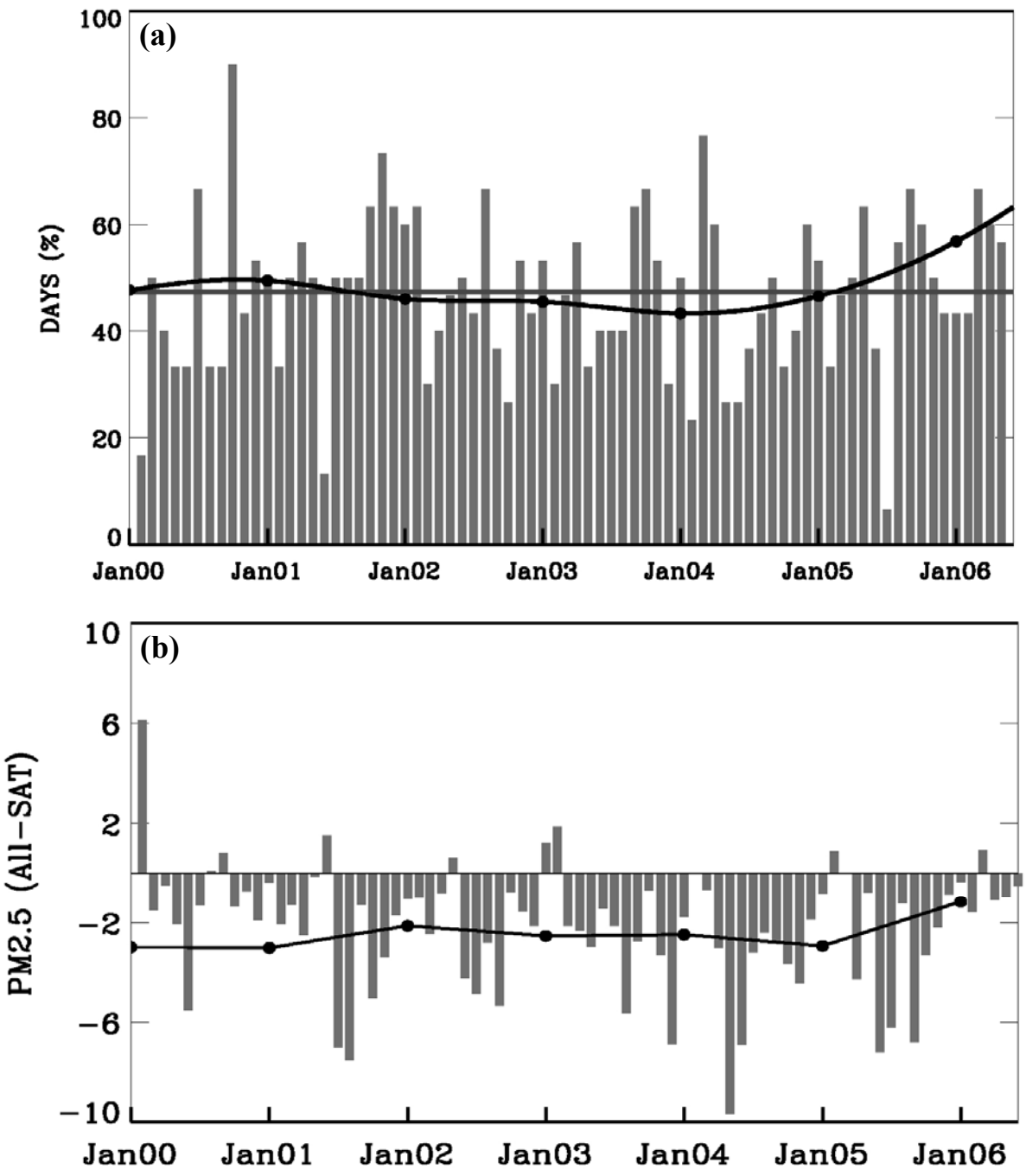

Figure 2.5 Time series analysis of difference in PM2.5 mass over NBHM site starting from February, 2000 to June 2006. (a) number of days (%) MODIS AOT data are available over NBHAM and (b) the monthly mean PM2.5 mass from all ground observation minus PM2.5 mass from only those days when MODIS AOT data are available. Solid line with filled circle shows yearly mean values. Horizontal thick black line in panel (a) is for all time mean values corresponding to 47.

low aerosol (or PM) loading over NBHM are being removed, possibly due to cloud cover. This trend is consistent over the entire study period.

There is only 8 out of 77 months, when this difference is positive, and 50% of them fall in the winter season. The mean difference is -2.2 μgm^{-3} with maximum negative value of -9.7μgm^{-3} during May 2004. The maximum positive difference of 6.1μgm^{-3} was obtained during February 2000 since Terra-MODIS started observations on February 24, 2000 and only 5-6 days of data available during this month. Averaging each month separately over the entire seven year time period we find that January has minimum negative difference of -0.5 μgm^{-3} whereas February has a positive difference of 0.6 μgm^{-3} Again, summer months have large negative differences compared to winter months, which corresponds to more number of available days in winter and less during summer months (Table 2.3). The solid line with dots in Fig 2.4(b) shows the yearly mean values, which are always negative. The maximum difference (-3.0 μgm^{-3}) in annual mean was in 2001 whereas minimum difference (-1.2 μgm^{-3}) was during 2006.

2.4.3 Regression Analysis between PM2.5 and MODIS AOT

Particulate matter air quality monitoring from satellite is based on the relationship between satellite derived AOT and surface measured PM2.5 mass concentration [*Wang and Christopher,* 2003*; Gupta et al.,* 2006]. The accuracy of satellite estimated PM2.5 over any given location depends on the robustness of the regression relationship between AOT and PM2.5 mass, which can depend on the accuracies of retrieved AOT and other factors such as vertical distribution of aerosols. Local meteorological conditions also govern the PM2.5 mass concentration and hence their inclusion in the AOT - PM2.5

Table 2.3 Regression coefficients, mean and standard deviation (σ) of PM2.5 and MODIS AOT using criteria number one.

	Slope	Intercept	R	Counts	PM2.5	σ	AOT	σ
Year								
2000	35.81	16.62	0.58	147	25.09	12.63	0.24	0.21
2001	28.95	16.39	0.50	181	22.02	10.95	0.20	0.19
2002	21.27	14.78	0.51	167	19.50	9.35	0.22	0.22
2003	22.36	15.43	0.50	165	19.88	9.65	0.20	0.22
2004	25.52	16.18	0.46	158	20.16	10.41	0.16	0.19
2005	32.13	16.33	0.57	169	22.67	11.71	0.20	0.21
2006	26.36	13.67	0.60	103	18.23	8.58	0.17	0.20
Mean	**27.48**	**15.63**	**0.53**	**156**	**21.08**	**-**	**0.20**	**-**
Month								
1	18.34	13.28	0.29	92	14.93	7.60	0.09	0.12
2	8.55	14.04	0.08	73	14.78	7.35	0.09	0.07
3	12.03	14.43	0.17	110	15.91	7.73	0.12	0.11
4	29.11	14.33	0.44	109	19.18	9.59	0.17	0.15
5	24.91	15.43	0.51	92	21.59	8.80	0.25	0.18
6	19.10	18.08	0.46	82	24.45	9.74	0.33	0.23
7	25.82	15.20	0.57	73	26.31	10.61	0.43	0.24
8	20.54	19.13	0.49	87	27.87	9.52	0.43	0.23
9	28.81	19.61	0.56	90	26.52	11.47	0.24	0.23
10	50.59	18.72	0.64	101	26.60	13.03	0.16	0.16
11	47.90	16.02	0.49	94	20.35	11.52	0.09	0.12
12	-14.34	17.98	-0.11	87	17.14	8.17	0.06	0.06
Mean	**22.61**	**16.35**	**0.38**	**91**	**21.30**	**-**	**0.20**	**-**

relationship can improve the overall accuracies in PM2.5 mass estimation from satellite observations [*Gupta et al.,* 2006]. However, we do not attempt to add meteorological parameters to the analysis for this research.

Computation of a single-variate least-squares linear regression relationship between MODIS AOT and PM2.5 is performed. We now discuss the change in regression coefficient such as slope (m), intercepts (c), linear correlation coefficient (r) and number of collocated points (N) as a function of month, season and year over NBHM. Changes in these parameters between hourly averaged and 24 h averaged PM2.5 are explored. Figure 2.6a presents the month to month and annual variations of MODIS AOT (primary y-axis) along with daily mean PM2.5 mass concentration (secondary y-axis). Both MODIS AOT and PM2.5 mass concentration follow similar trends on monthly and yearly time scales as noted in section 4.1. Figure 2.5b shows hourly averaged (red lines) PM2.5 during Terra MODIS overpass time from 2000 to 2006 along with 24 h averaged PM2.5 (blue lines). Both hourly and 24 h values follow similar variations on monthly and yearly scales, but hourly averaged values are smaller compared to 24 h averaged values. The mean ratio of monthly mean obtained using 24hour averages to mean obtained using hourly average PM2.5 is 1.66±0.47.

The linear correlation coefficient between 24 h mean PM2.5 mass concentration and MODIS AOT is 0.52 with the regression relationship given by Equation (2.1).

$$PM2.5 = 15.8 + 27.5 \times MODIS\ \ AOT \cdots\cdots\cdots\cdots\cdots (2.1)$$

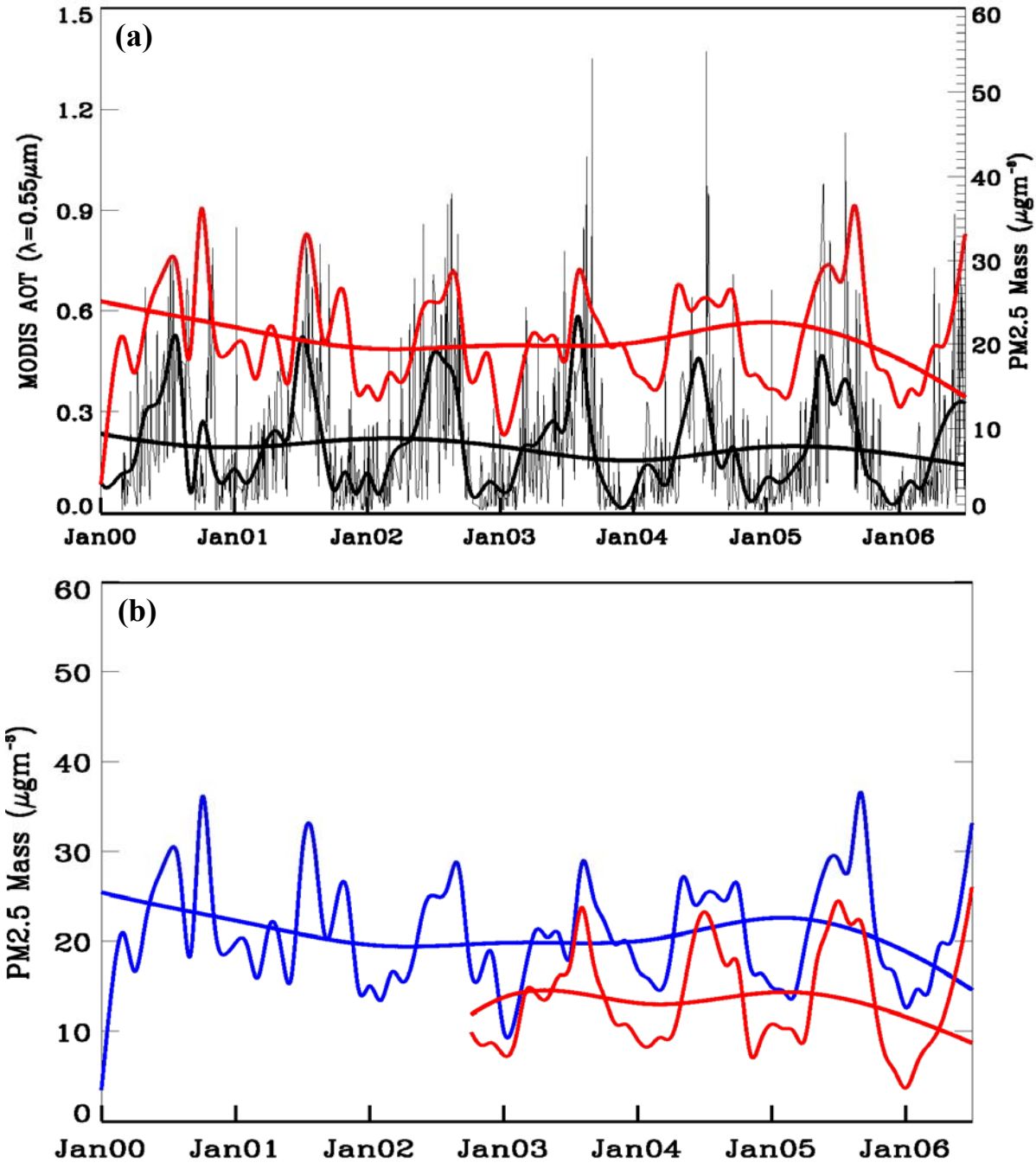

Figure 2.6 Trend analysis of surface measured PM2.5 mass (1 hour and 24 hour) and MODIS AOT over the station. (a) daily, monthly and annual mean AOT as time series (black lines) and monthly and annual mean PM2.5 mass concentration (red lines). (b) monthly and annual mean PM2.5 mass obtained from 24 hour mean PM2.5 mass data (blue lines) along with monthly and annual mean PM2.5 mass obtained from 1 hour (during satellite overpass) PM2.5 data (red lines).

When a similar analysis is performed on hourly averaged PM2.5 data, the correlation increases to 0.62 and the corresponding regression equation becomes Equation (2.2).

$$PM2.5 = 8.8 + 29.4 \times MODIS\ \ AOT \cdots\cdots\cdots\cdots\cdots (2.2)$$

These regression equations can be used to estimate the ground level PM2.5 mass concentration. As expected, the instantaneous MODIS AOT correlate better with hourly averaged PM2.5 compared to 24 h averaged PM2.5 due to diurnal variations in PM2.5 mass measurements. National Air Quality Standards are set for 24 h mean PM2.5 mass concentration and it is an important parameter to monitor, hence further analysis is focused only on 24 h mean PM2.5 and despite its moderate correlation, these relationship can estimate daily air quality categories with high degree of accuracy [*Wang and Christopher et al.,* 2003].

Several studies have shown that the relationship between PM2.5 and AOT varies substantially with location and time [*Engel-Cox et al.,* 2004*; Gupta et al.,* 2006]. To further explore this variability, regression coefficients for each month and year are calculated and presented in Table 2.3. Yearly analysis shows that value of *r* over different years does not vary significantly and ranges between 0.46 in 2004 and 0.60 in 2006. Compared to yearly variations, *r* demonstrates more variation from month to month reflecting its dependence on season. Except for December, the correlation is positive and varies from minimum of 0.08 to maximum of 0.64. In December *r* = -0.11 and corresponds to very low AOT values (0.06). The negative correlation may be associated with large uncertainties in MODIS AOT, which can be more than 70% for AOT values

less than 0.1 over land [*Remer et al.,* 2005*, Levy et al.,* 2007a]. These low AOT's correspond to low PM2.5 concentrations and in general, summer and fall correlations are higher compared to winter and spring. Highest correlations are associated with mean high AOT and PM2.5 mass concentration, which is consistent with our previous analysis in other parts of the world [*Gupta et al.,* 2006; 2007].

Intercept in regression equation represents the background levels of PM2.5 mass concentration in the absence of AOT (zero AOT). In other words, intercept represents the minimum level of particle concentration for which satellite derived AOT is sensitive. Below the level of intercept, satellite signals are weak and detection of aerosols is difficult. Value of intercept is relatively constant as a function of time and only changes between 13.7 and 16.6 μgm^{-3} with mean value of 15.6 μgm^{-3}. In general, the intercept is greater in summer and fall months compared to winter and spring, which is also associated with high mean values of PM2.5 mass in respective months. The slope of this relationship is an important parameter for converting AOT into surface PM2.5 mass. The slope changes from 21.27 to 35.81 over different years with mean value of 27.48. The variations in monthly slope values are large compared to yearly values and even becomes negative (-14.34) during December. Slope values could also be dependent on the local meteorological conditions as well as vertical distribution of the aerosols. Relative humidity could be an important parameter to observe while deriving these relationships because same dry PM2.5 mass concentration under different relative humidity condition can produce different AOT due to changes in scattering properties of aerosols.

2.4.4 MODIS AOT, Box Size, and Quality Flags

Satellite aerosol retrieval algorithms convert the measured reflectance to a geophysical parameter related to aerosol concentration known as AOT. There are many quality flags associated with the input data, processing methods and output data quality, which are reported in the MOD04 AOT product. The MODIS science team recommends using these quality flags along with actual AOT values for research studies. Depending on the research study, the quality flag restriction could be relaxed or stringent. In the current study, the changes in mean AOT values due to various combinations of quality flags relevant for particulate matter air quality research are examined. The effect of box size (number of pixels in spatial collocation) around the ground station on mean AOT values is quantitatively estimated for three different box sizes. Table 2.2 presents all nine criteria used to evaluate the change in mean AOT values at NBHM on daily, monthly and annual basis. The following discussion is separated into two parts; first, the effect of box size on mean AOT value is evaluated and then the effects of using different quality flags are discussed.

2.4.4.1 Varying box size around ground station

Criteria numbers 1 to 3 (CN1 to CN3) in Table 2.2 use all retrieved AOT values to obtain mean AOT value over NBHM without any quality flag restrictions, but for three different box size of 5x5 (0.5x0.5 degrees, AOT5), 4x4 (0.4x0.4 degrees, AOT4), and 3x3 (0.3x0.3 degrees, AOT3) pixels. Decreasing box size, decreases the number of pixels used to derive MODIS AOT values for NBHM, and may affect the overall mean AOT and these can change PM2.5 estimation from satellites due to change in spatial sampling

of aerosols [*Hutchison et al.,* 2005]. Larger box sizes generally indicates sampling of more heterogeneous type of aerosols, which can over or under estimate AOT values compared to point measurements from ground stations. *Ichoku et al* [2002] reported that a 5x5 box is a good approximation for AOT validation with 1 hour averaged AERONET AOT values, which is supported by arguing that average speed of aerosol transport in mid-troposphere is about 50km/hr based on analysis of TOMS aerosol index images over the Atlantic Ocean.

However, the current focus of the study is AOT for air quality applications, which is measured at surface in the form of PM2.5 mass concentration. Since the average travel speed of aerosols (air mass) near the surface (lowest few hundred meters) is much less than mid-troposphere, one hour averaged PM2.5 measurements may not represent the air mass in the 50x50 km area sampled by MODIS instrument. As a result, a smaller 3x3 box size is considered while evaluating the effects of quality flags in second part of this section. Reducing box size further might result in a statistically insignificant number of pixels and may introduce additional uncertainties. Figure 2.7 presents differences in daily, monthly, and annual mean AOTs due to changes in box sizes. The inset in figure 2.7 contains the frequency distribution of differences in AOT4 and AOT3 values from AOT5. The difference is larger between AOT5 and AOT3 compared to AOT5 and AOT4. Daily mean differences of AOT5-AOT4 and AOT5-AOT3 are -0.004 and -0.010 with ranges of -0.1 to 0.11 and -0.19 to 0.23 respectively for the entire time period. No specific trends or seasonal and annual time scales were observed when varying box sizes. Frequency distribution also shows that the difference in AOTs between -0.03 and 0.03 is

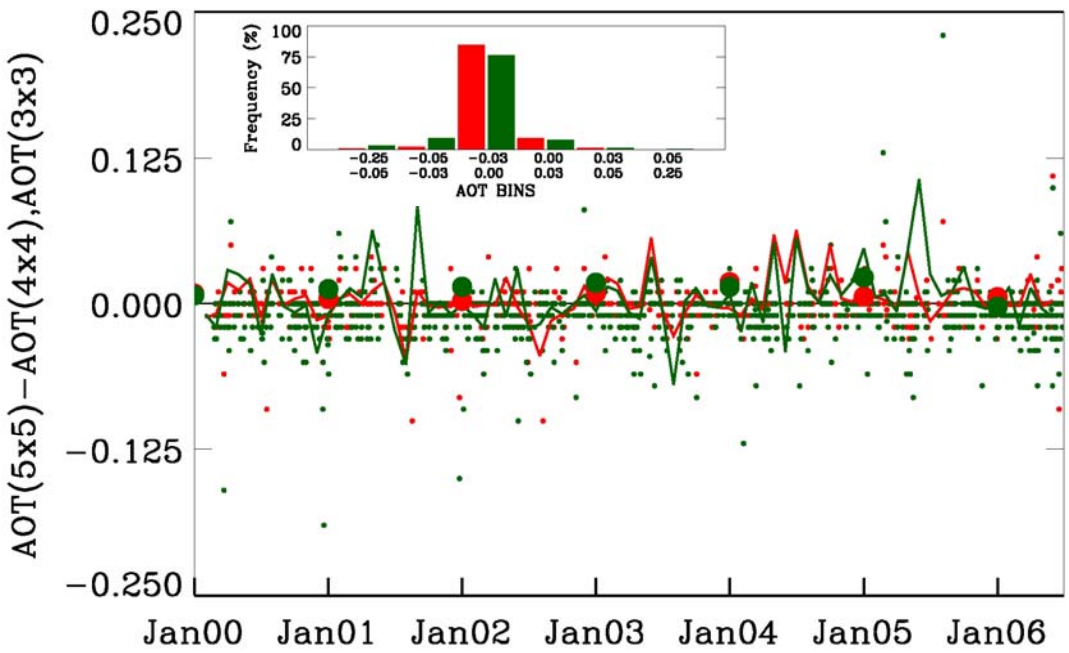

Figure 2.7 Time series analysis of difference in AOTs due to change in box size around the ground station. Y-axis represents AOT5-AOT4 (red color in plot) and AOT5-AOT3 (green color in plot). Big filled circles are yearly mean, solid lines are monthly mean and small filled circles are daily AOT values. The inset panel shows frequency distribution of difference in AOTs for daily values.

greater than 90% of the time that is well within uncertainty limits of MODIS AOT retrievals [*Levy et al.,* 2007a, b]. Further investigation reveals that the mean of AOT5 values for which the difference is between -0.05 to 0.05 is less than 0.3 whereas it is greater than 0.45 for differences larger than 0.05.

Therefore, it is clear that during high aerosol loading (AOT>0.3) the differences in instantaneous AOT due to changes in box size could be significant while the differences are within retrieval uncertainties when aerosol loading is low (AOT<0.3). AOT values do not change as a function of box size up to second digit of significance (Table 2.4). It is also important to note that, reducing box size from 5x5 to 3x3 reduces the number of available data by about 8%. The regression coefficients for the AOT - PM2.5 relationship (see section 2.3) are recalculated and we found that they do not change significantly as a function of box size and the number of data points. Slope values changed by less than 7%, intercept by less than 0.5% and linear correlation coefficient improved by less than 2%.

2.4.4.2 Applying Quality Flags

Criteria numbers 3 to 9 (CN3 to CN9) in Table 2.2 use a constant box size of 3x3 pixels around the PM2.5 station and only quality flags associated with MODIS AOT retrievals over land are changed. The primary focus of this analysis is to determine importance of the quality flags for daily, monthly and yearly aerosol analysis. We examine how much bias or uncertainty can be introduce in the AOT values if one does not use any quality flags to represent the aerosol loading. Criteria number 9 is the most stringent in terms of quality flags and uses only good or very good retrievals,

Table 2.4 Effect of box size on AOT values at daily, monthly and yearly scales.

Daily					
Box	**Min.**	**Max.**	**Mean**	**Std. Dev.**	**Days (%)**
AOT5	0.010	1.370	0.198	0.205	45.049
AOT4	0.010	1.370	0.190	0.201	41.635
AOT3	0.010	1.410	0.185	0.197	37.000
Monthly					
AOT5	0.016	0.583	0.200	0.136	47.359
AOT4	0.019	0.610	0.196	0.138	43.680
AOT3	0.019	0.652	0.195	0.139	38.571
Yearly					
AOT5	0.156	0.235	0.196	0.027	42.818
AOT4	0.138	0.226	0.188	0.030	39.491
AOT3	0.142	0.228	0.184	0.027	34.873

which are identified as land pixels with a solar zenith angle less than 60 degree under less than 30% cloud cover conditions. Mean AOT using CN9 are considered as best AOT (BAOT) value and all other criterions (CN3 to CN8) are evaluated with respect to CN9 (Table 2.5). A negative difference represents an overestimation in AOT. Differences in daily (instantaneous) AOT values range from -0.11 to 0.08 while mean value varies from one criterion to other. Table 2.5 demonstrates that the mean difference is always negative in daily, monthly and yearly analysis, which indicates an overestimation of AOT values when quality flags are not used. Figure 2.8 indicates that AOT values on monthly and yearly scales are generally overestimated compared to BAOT. CN4 and CN5 represent two different cases with very minor differences. CN4 does not include any cloud flags whereas CN5 restricts cloud cover above 90%. AOT retrieval process and screening of unwanted pixels are such that most of the time CN4 and CN5 will have same AOT value. Differences in mean AOT values while using different criteria on daily basis are very small. The differences are larger in monthly and yearly mean values, which could arises due to sampling issues discussed in the previous section. Table 2.6 presents these differences for each month separately to shows seasonal variability in AOT for different criteria. Overall, there is not much seasonal dependence, but summer months (May to July) have the maximum difference while using all criteria, which could be associated with changes in cloud cover.

Figure 2.9 presents the reanalysis of regression equations as discussed in section 4.3 with application of quality flags on AOT. The main objective of this analysis is to understand the differences in regression coefficients that can occur due to the use of different quality flags.

Table 2.5 Effect of quality flags on mean AOT values over the stations. Daily, monthly and yearly differences (BEST-AOT).

Daily			
Difference	**Min.**	**Max.**	**Mean**
CN9-CN3	-0.0800	0.0800	-0.0028
CN9-CN4	-0.1100	0.0800	-0.0032
CN9-CN5	-0.1100	0.0800	-0.0032
CN9-CN6	-0.1100	0.0800	-0.0031
CN9-CN7	-0.0700	0.0800	-0.0022
CN9-CN8	-0.1100	0.0700	-0.0004
Monthly			
CN9-CN3	-0.1120	0.1333	-0.0149
CN9-CN4	-0.1158	0.1630	-0.0115
CN9-CN5	-0.1158	0.1630	-0.0115
CN9-CN6	-0.1158	0.1630	-0.0102
CN9-CN7	-0.0606	0.1450	-0.0034
CN9-CN8	-0.0545	0.0783	-0.0031
Yearly			
CN9-CN3	-0.0411	-0.0006	-0.0182
CN9-CN4	-0.0426	0.0003	-0.0179
CN9-CN5	-0.0426	0.0003	-0.0179
CN9-CN6	-0.0380	-0.0017	-0.0172
CN9-CN7	-0.0199	-0.0004	-0.0105
CN9-CN8	-0.0124	0.0017	-0.0041

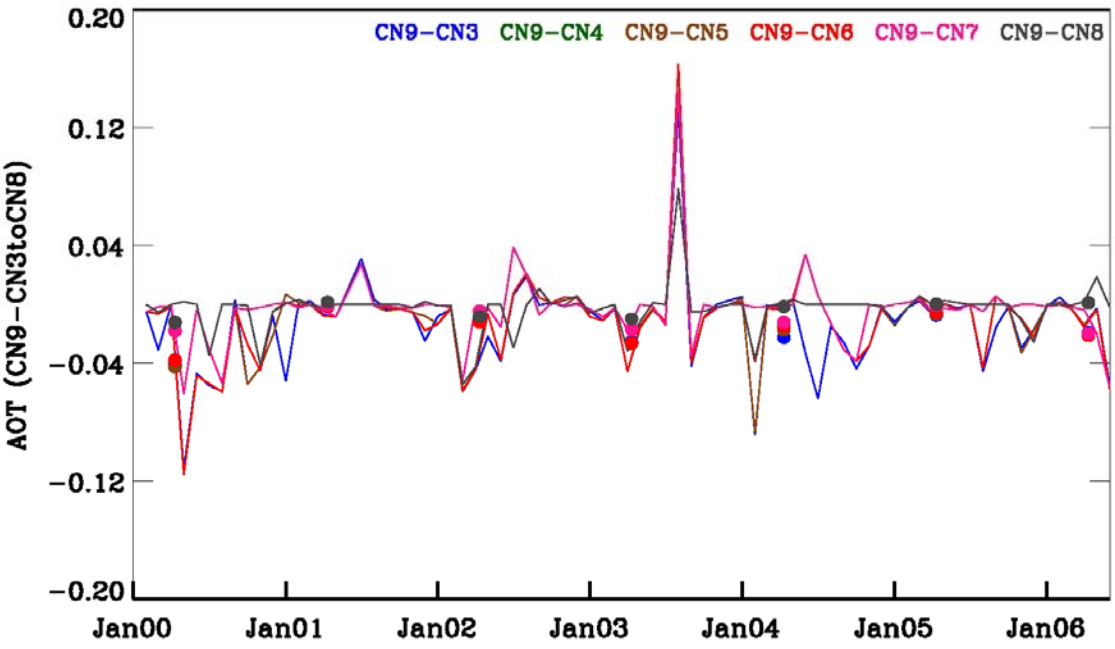

Figure 2.8 Time series analysis of difference in AOTs after applying quality flags. Y-axis represents Best AOT (CN9) - all other AOTs (CN3 to CN8). Big filled circles are yearly mean, solid lines are monthly means.

Table 2.6 Difference in AOTs for each criterion averaged over each month separately over entire time period.

Month	**Difference**					
	CN9-CN3	**CN9-CN4**	**CN9-CN5**	**CN9-CN6**	**CN9-CN7**	**CN9-CN8**
1	-0.0122	-0.0045	-0.0045	-0.0058	-0.0006	-0.0034
2	-0.0149	-0.0151	-0.0151	-0.0087	-0.0025	-0.0057
3	-0.0131	-0.0097	-0.0097	-0.0095	-0.0079	-0.0079
4	-0.0171	-0.0174	-0.0174	-0.0177	-0.0051	-0.0106
5	-0.0225	**-0.0210**	**-0.0210**	**-0.0208**	**-0.0134**	0.0021
6	-0.0298	-0.0196	-0.0196	-0.0194	-0.0077	0.0001
7	**-0.0155**	-0.0041	-0.0041	-0.0041	0.0045	**-0.0108**
8	*0.0060*	*0.0112*	*0.0112*	*0.0112*	*0.0155*	*0.0129*
9	-0.0141	-0.0112	-0.0112	-0.0108	-0.0117	0.0010
10	-0.0182	-0.0175	-0.0175	-0.0131	-0.0075	-0.0009
11	-0.0179	-0.0188	-0.0188	-0.0143	-0.0013	-0.0089
12	-0.0080	-0.0073	-0.0073	-0.0072	-0.0003	-0.0039
Mean	**-0.0148**	**-0.0113**	**-0.0113**	**-0.0100**	**-0.0032**	**-0.0030**

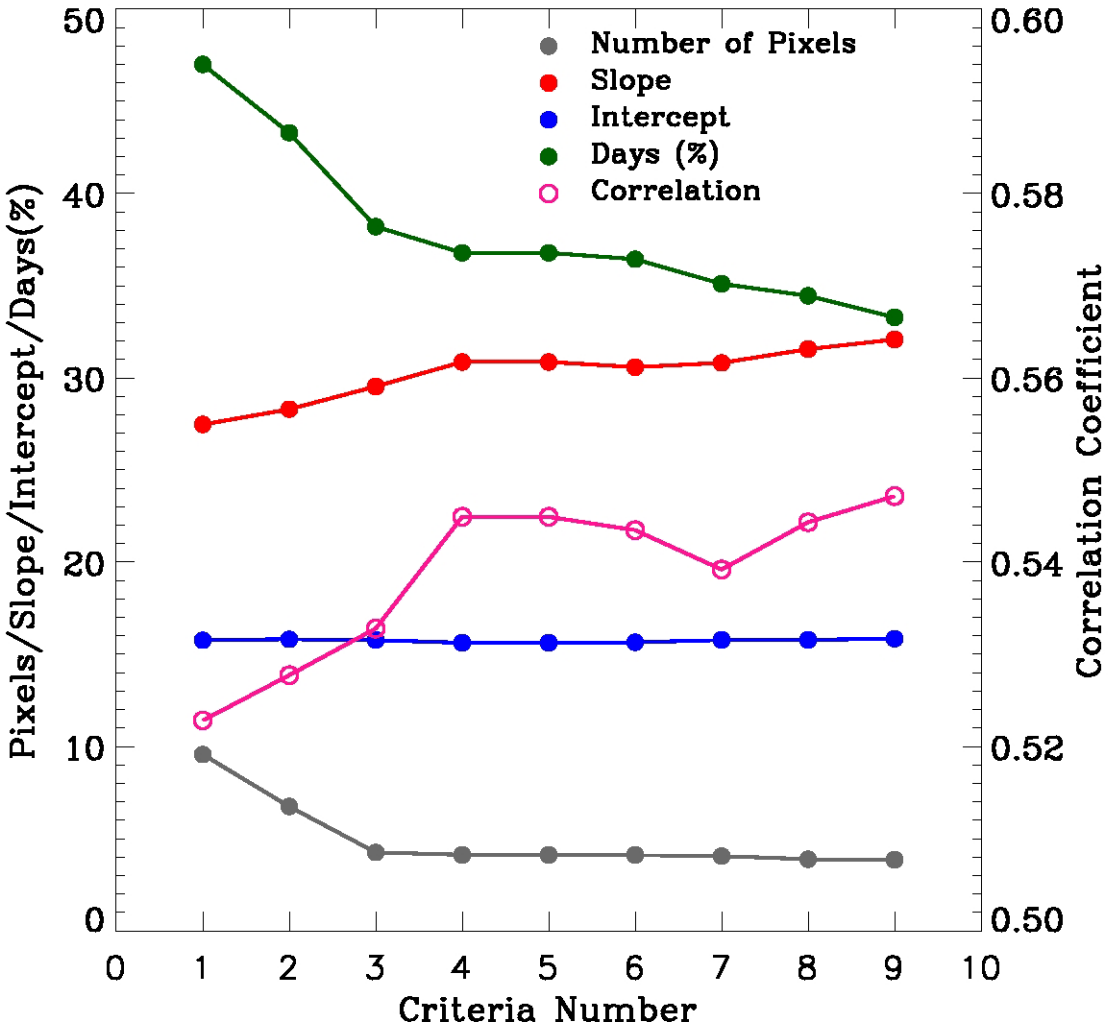

Figure 2.9 Revisiting regression analysis using AOT from 9 different criteria as shown in table 2.2. Parameters plotted here are number of AOT pixels for averaging over ground station, slope, intercept and correlation coefficient for regression line between PM2.5 and AOT and number of day (%) the AOT data available.

The effect on mean number of pixels is only visible from changing box size, and it decreases as box size reduces from CN1 to CN3. We only plot all time mean values, but there could be differences in the number of pixels due to changes in quality flags over different time periods. The changes in number of pixels while using CN3 to CN9 are mainly associated with changes in cloud cover. Slope generally increases from 27.47 to 32.08 for CN1 to CN9. This could be due to the decrease in mean AOT value from CN1 to CN9. Intercept is almost constant with mean value of 15.6, which represents a constant background PM2.5 loading over the station through out the study period. The number of days when data were available decreased from 47.0 % for CN1 to 33.3 % for CN9. However, correlation between PM2.5 and AOT does not vary significantly and only improves from 0.52 to 0.55 from CN1 to CN9.

2.5 Summary

Seven year surface measured PM2.5 mass concentration and MODIS-Terra derived aerosol optical thickness data sets were collected over one location in Southeastern United States (NBHM) to assess different aspects of satellite monitoring of particulate matter air quality. Surface and satellite data were analyzed to asses the long term air quality trends, availability of satellite data for air quality applications, the relationship between surface measured PM2.5 mass concentration and satellite derived integrated columnar aerosol loading (AOT) with the new generation of Collection 5 MODIS aerosol products, application of quality control flags on aerosol optical thickness values over the station, and effects on these flags on the PM2.5 - AOT relationships.

Our results indicate that NBHM air quality has improved from 2000 to 2006 with unhealthy conditions during summer when compared to other seasons. Since satellites can provide particulate matter air quality information only during cloud-free and favorable surface conditions we calculated the availability of satellite data for this station. A significant conclusion of this study is that even though NBHM is sampled only 50% of the time by Terra-MODIS, the monthly, seasonal and yearly means are different only by $2\mu gm^{-3}$ when compared to using a 100% data availability rate from ground measurements. Our analysis of quality control flags and box sizes for evaluating PM2.5-AOT relationships indicate that for daily analysis using quality control flags are critical but for monthly and yearly analysis these flags may not be necessary. Finally, quality flags also do not affect the PM2.5 - AOT relationship significantly but reduces the available satellite data by up to 14% from all retrievals to best retrievals. Hence depending on the requirement of sample size as well as accuracies, quality flags can be used or ignored. These results indicate that satellite data can be a powerful tool for evaluating air quality in regions especially when ground measurements are not available. Direct extrapolation of results from this study to other region may not be possible due to change in local conditions and satellite retrievals.

CHAPTER 3

AIR QUALITY ASSESSMENT USING MULTIPLE REGRESSIONS APPROACH

3.1 Introduction

Monitoring and forecasting of surface level particulate matter (PM) pollution is one the top priority for many environmental agencies around the world due to its adverse effects on living biota. A medical study by *Pope III et al*, [2002] concluded that fine particles and sulfur oxide related pollutants are associated with lung cancer and cardiopulmonary mortality. The same study also states that an increase of 10 μgm^{-3} in fine particulate (PM2.5, particles less than 2.5 µm in aerodynamic diameter) can cause a 4 - 8% increase in cardiopulmonary, and lung cancer mortality.

Typically PM2.5 is monitored from the ground and these surface monitoring stations do not provide adequate spatial sampling. In recent years, the potential of using satellite measurements of aerosols for monitoring surface PM2.5 mass over various regions has been explored [*e.g. Wang and Christopher*, 2003].

Several research papers have outlined the methods by which different satellite products can be used to obtain surface PM2.5 [*e.g. Wang and Christopher,* 2003*; Engel-Cox et al.,* 2006*; Hutchison et al.,* 2005*; Gupta et al.,* 2006*, Liu et al.,* 2004*; van Donkelaar et al.,* 2006]. In summary, first the columnar satellite-derived aerosol optical thickness (AOT) values are related to surface PM2.5 mass measurements. Then this AOT-PM2.5 relationship is used to convert the satellite measurements to PM2.5 mass concentration and then to air quality indices based on EPA guidelines [*e.g. Al-Saadi et al,* 2005]. However most studies have concluded that the PM2.5-AOT relationship alone cannot be used to estimate surface level particulate matter since the vertical distribution of aerosols and other meteorological parameters such as humidity and temperature could also be important [*Liu et al.,* 2005]. Since the satellite measurements provide columnar retrievals relating this to surface values requires information about the vertical distribution of aerosols. If aerosols are transported aloft and the satellite retrievals have recorded values for AOT this does not necessarily mean that the ground monitors capture that event. In this case relating the AOT to PM2.5 values is not meaningful.

The vertical distribution of aerosols can be inferred from ground based [*Fernald,* 1984*; Ansmann et al.,* 2000] and from space-borne LIDARs such as CALIPSO [*Winker et al.,* 2003]. Radar data could also be useful for estimating plume injection heights [*Jones and Christopher,* 2008]. However there are various limitations with these data sets. The swath width of space borne lidars such as CALIPSO is very narrow (about 70 km) and global coverage is only achieved over several weeks. Ground-based lidars on the other hand are sparse in spatial coverage and do not operate on a continual basis. Using limited observations of vertical distribution of aerosols from lidars, some studies have

demonstrated that aerosols are well-mixed and mostly confined within the planetary boundary layer (PBL) [*Ansmann et al.,* 2000]. Thus, the height of PBL (HPBL) or the day time mixing layer height can be used as a good surrogate to the height of the aerosol layer. This can be used to scale the column AOT value into boundary layer extinction value [*Liu et al.,* 2006] to improve the AOT-PM2.5 relationships. Other parameters such as humidity could also affect this AOT-PM2.5 relationship since aerosols grow as a function of humidity. Therefore, in this paper we assess whether the use of meteorological parameters will improve the relationship between AOT and PM2.5. We construct simple AOT-PM2.5 relationship that we call two-variate method (TVM) and then compare this to a multi-variate method (MVM) that utilizes various meteorological parameters.

3.2 The AOT-PM2.5 Assessment using Meteorology

Fine particular matter in the atmosphere is produced by gas to particle conversion mechanism as well as being directly injected from various sources due to various anthropogenic and natural activities. The meteorological conditions that strongly influence the concentration of PM2.5 particles include temperature, relative humidity and height of planetary boundary layer [*Pandis and Seinfeld,* 2006]. Other processes that impact particulate matter concentration includes small to large scale transport by winds, horizontal and vertical dispersion, variations in available sun light for photochemical reactions due to clouds and seasons, temperature gradients, available moisture, and most importantly the dilution of pollution in atmospheric boundary layer due to changes in vertical mixing. The variability in these meteorological conditions primarily governed by

the movement of large scale high and low pressure systems, diurnal heating and cooling, and topography. Temperature can enhance the photochemical reactions in the atmosphere and hence production of PM2.5 particles. Temperature inversion can also affect the strength of emission source. It can also reduce the vertical mixing and therefore increase chemical concentration of precursors. Higher concentration of precursors produces faster and more efficient chemical processes that convert gaseous emissions into particles. High relative humidity can enhance the growth and production of secondary particles and hence change in the size distribution of particles as well as change in optical properties by modifying scattering efficiencies can be observed [*Wang et al.*, 2007].

Dilution of pollution due to change in boundary layer height (HPBL) and moisture contain are of significant importance in satellite remote sensing of particulate matter. PM2.5 mass concentrations measured at the surface will be low during conditions of high HPBL compared to condition of small boundary layer heights whereas satellite will measure almost same columnar AOT. Figure 3.1 presents a schematic of two different cases of temperature inversion, HPBL, and their influence on surface measured PM2.5 mass concentration. In the first case (Figure 3.1, top panel), vertical temperature profiles shows weak and high inversion, which is easily broken due to solar heating at sunrise and the boundary layer starts growing and reaches to a maximum height during the afternoon. As a result of increase in HPBL, existing pollution level dilute in the growing boundary layer and hence surface station will measure reduced PM2.5 mass concentration values. In the second case (Figure 3.1, bottom panel), strong and low inversion does not allow growth of HPBL due to solar heating, therefore slowing down the process of dilution and the surface stations measure high PM2.5 mass concentrations

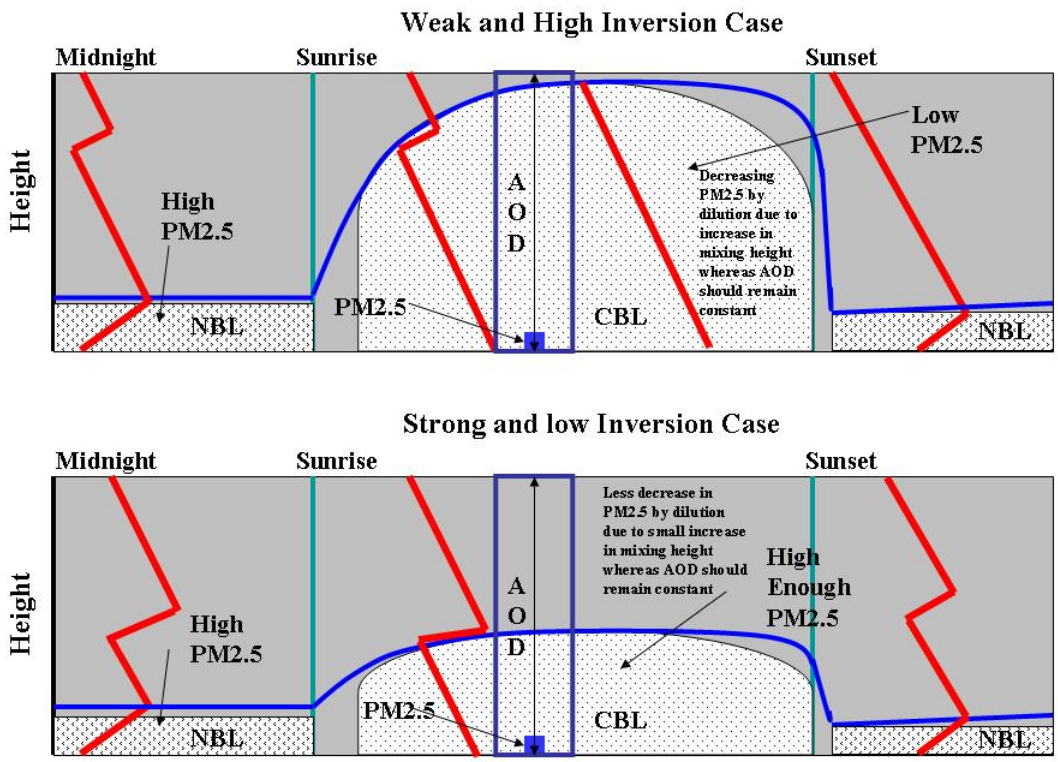

Figure 3.1 Schematic showing evaluation and growth of planetary boundary layer height under weak and strong temperature inversion conditions.

when compared to the first case. In both cases, space-borne measurements will still measure columnar loading of the aerosols and hence theoretically the AOT should remain constant. Therefore, accounting for proper HPBL in estimation of PM2.5 is important when using satellite-retrieved AOT as a surrogate.

3.3 Data and Study Area

To assess surface level particulate matter, three years of (2004-2006) hourly PM2.5 mass concentrations from the ground stations, Terra-MODIS AOT at 0.55µm and meteorological fields from hourly RUC reanalysis over 85 AirNow stations in south east United States are used (Figure 3.2). Table 3.1 provides details on the different data sets.

3.3.1 Surface PM2.5 Mass

The United Stated Environmental Protection Agency (EPA) and its state partners maintain several air quality monitoring networks in the United States. These networks monitor the mass concentration of particulate air pollutants at the ground. PM2.5 data from these networks include 24-H average (daily) and hourly PM2.5 mass concentrations. The PM2.5 mass measured in μgm^{-3} is most relevant to the present study. This study uses both hourly and daily mean PM2.5 mass concentration data sets from 85 ground stations in south east United States as shown in Figure 3.2**.** $PM_{2.5}$ mass concentration over these stations is measured using a Tapered-Element Oscillating Microbalance (TEOM) instrument with an accuracy of ±1.5 μgm^{-3} for hourly averages. Hourly average PM2.5 mass concentration values are used to derive air quality categories whereas daily mean values are used to monitor and assess the air quality.

Table 3.1 Detail information on various surface, model and satellite data sets used for particulate matter air quality assessment in south east United States.

No.	**Parameters**	**Sensors**	**Temporal Resolution**	**Resolution**	**Source**
1	PM2.5	TEOM	Hourly	Point measurements	EPA's AirNow-Tech
2	AOT, CF	MODIS-Terra Satellite (MOD04)	Instantaneous	10x10 km	Level 1 and Atmosphere Archive and Distribution System
3	Meteorological Fields	RUC reanalysis	Hourly	20x20 km	Atmospheric Radiation Measurement (ARM) data archive

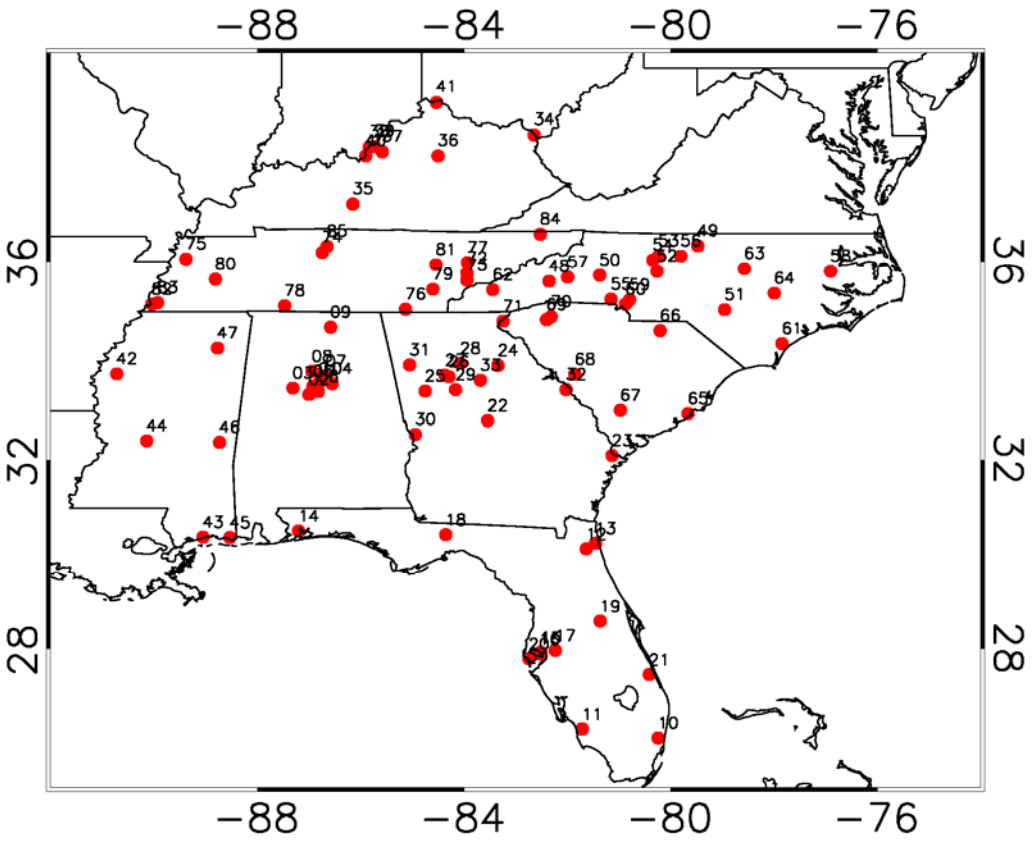

Figure 3.2 Study area showing major cities in the area along with locations of PM2.5 monitoring stations under EPA AirNow network.

3.3.2 Satellite Aerosol Data

The MODIS onboard NASA's Terra (equatorial crossing time of 10:30 a.m.) and Aqua (equatorial crossing time of 1:30 PM) satellites provide systematic retrieval of cloud and aerosol properties over land [*King et al.,* 1999]. MODIS provides spectral information of aerosol optical properties in seven different wavelengths over ocean and in three wavelengths over land [*Levy et al.,* 2007]. The AOT, representing the columnar loading of aerosols in the atmosphere, is an important aerosol parameter retrieved from satellite observations. Recently, a new version of the MODIS algorithm (collection 5, V5.2) has replaced an older version (collection 4, V5.1). The new algorithm uses new aerosols model and improved estimation of surface reflectance. This algorithm applies stringent criteria to select appropriate pixels in the retrieval process thereby reducing the total number of data points [*Levy et al.,* 2007]. Several validation studies over AERONET locations conducted over land reveal that 57% of MODIS AOT retrievals (Collection 4) are within expected uncertainty levels of ±0.05±0.15*AOT [*Remer et al.,* 2005]. Preliminary results from a validation exercise of MODIS collection 5 shows that 72% of the retrievals fall within expected uncertainty over land [*Remer et al.,* 2008], which shows an improvement over collection 4 data sets.

3.3.3 Meteorology from RUC Analysis

The Rapid Update Cycle (RUC) is an operational atmospheric prediction and assimilation system comprised primarily of a numerical forecast model and an analysis system to initialize that model. The RUC has been developed by the Earth System

Research Laboratory at NOAA to serve users needing short-range weather forecasts [*Benjamin et al.,* 2004]. RUC runs operationally at the National Centers for Environmental Prediction (NCEP). A new version of the RUC has been implemented at the NCEP with an improved horizontal resolution (20km), increased number of vertical computational levels (50 levels, typically from surface to 45-60 hPa) and improvements in the analysis and model physical parameterizations. The primary goal of the development of the 20-km RUC (or RUC20) has been the improvement in warm-season and cold-season quantitative precipitation forecasts. The RUC20 provides improved forecasts for these variables, as well as for wind, temperature, and moisture [*Benjamin et al.,* 2002]. Hourly analysis data of air temperature at 2 meter height (TMP), surface relative humidity (RH), wind speed at 10 meter (WS), and height of planetary boundary layer (HPBL) at 20x20 km^2 spatial resolution are used in this study. HPBL is one of the important parameters in evaluating air quality from satellite data. It is a diagnostic variable in the RUC reanalysis and it is calculated using vertical profiles of virtual potential temperature. We use this as a surrogate for aerosol height since other measurements are not available over the entire area of study.

3.4 Development of Multiple Regression Equations

Integrating satellite, surface and meteorological parameters derived from models at the same temporal and spatial scales is one of the important steps towards development and analysis of a statistical model. The integration of these different variables is performed in two steps. First, satellite data is obtained for each PM2.5 ground location, and secondly RUC20 data is obtained for each satellite-PM2.5 data match-up. In our

previous study [*Gupta and Christopher*, 2008a], we have shown that averaging MODIS AOT over a box size of 0.5x0.5 deg (~5x5 pixels) centered around PM2.5 station is most appropriate for this type of analysis. We follow the same methodology outlined in *Gupta and Christopher* [2008a] to obtain MODIS AOT and other relevant parameters over each PM2.5 station. Further screening of the data is done by eliminating all those AOT-PM2.5 pairs where numbers of pixels are < 3 and standard deviation in AOT is > 0.5. This standard deviation is set to avoid possible cloud contamination in AOT data. We use hourly measurements of PM2.5, matched with the MODIS Terra AOT closest to the satellite overpass time. The meteorological parameters are also obtained for each AOT-PM2.5 data point by using same spatial and temporal approach used to match AOT-PM2.5 data. The matched dataset contains spatio-temporally collocated satellite derived AOT, cloud fraction (CF), number of pixels (MPIX) used to average AOT values around the PM2.5 station, surface measured PM2.5 mass concentration (PM2.5) and RUC20 derived meteorological fields for 85 PM2.5 monitoring stations in EPA region 4. The final dataset contains 32,834 collocated samples, which are used in development, testing and validation of the statistical model. To produce the final dataset (Satellite-Surface-Model (SSM) now onwards), almost 1TB of combined RUC, MODIS and PM2.5 data has been processed.

Figure 3.3 provides a flow chart of the methods. First, we developed a simple two variate method (TVM) equation (1), where MODIS AOT is used to estimate surface level PM2.5 mass concentration. Then, meteorological parameters are added to the analysis to form multiple linear regression (MVM) equations (2) to estimate PM2.5 mass concentration.

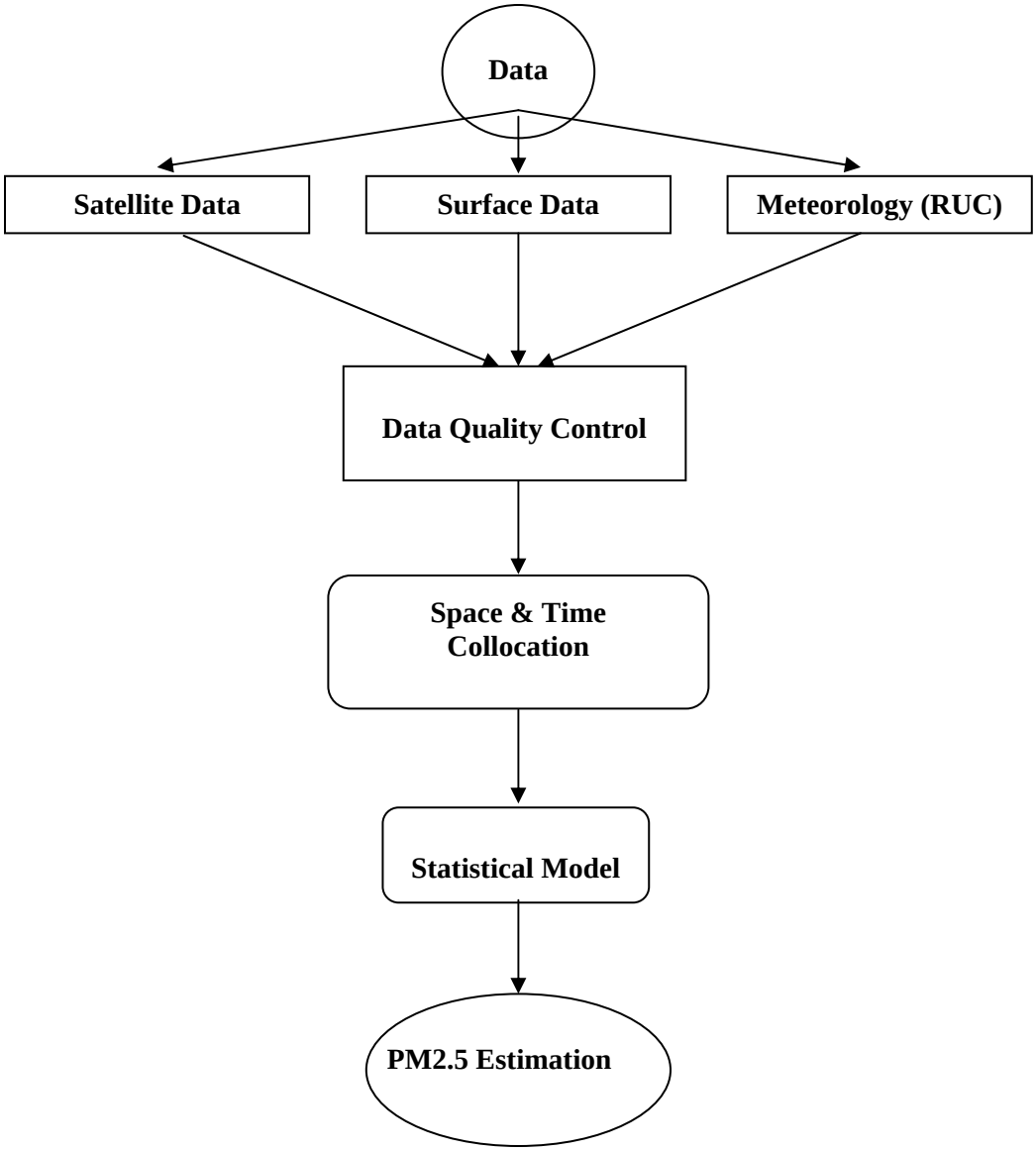

Figure 3.3 Schematic showing different data sets and framework of model to integrate different data sets for particulate matter air quality.

Regression coefficients were calculated using SSM data sets for equation 3.1 and 3.2 and then these equations are used to calculate PM2.5 mass concentration using input parameters from satellite and RUC meteorological fields. The general form of TVM model is shown in equation 3.1 and the MVM model in equation 3.2:

$$PM_{2.5} = C + M \times AOT \cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots (3.1)$$

$$PM_{2.5} = C_1 + C_2 \times AOT + C_3 \times TMP + C_4 \times RH + C_5 \times HPBL + C_6 \times WS + C_7 \times CF \cdots (3.2)$$

Where PM2.5 is PM2.5 mass concentration (μgm^{-3}), AOT is MODIS AOT at 550 nm (unit less), C is the intercept and M is slope for the TVM. C_1 is intercept for the MVM whereas C_2-C_7 are regression coefficients for predictor variables including AOT, temperature (K), relative humidity (%), height of planetary boundary layer (m), wind speed (m/s), and cloud fraction (%) respectively.

To obtain an overview of the parameters used in these two methods, the frequency distribution of all variables is shown in Figure 3.4. The frequency distribution of AOT and PM2.5 are similar, indicating that AOT contains information related to PM2.5 mass. All of the variables show significant seasonal variability. The mean MODIS AOT value is 0.19 and the corresponding PM2.5 mass concentration has a mean value of 13.5 μgm^{-3}. The AOT values during summer are the highest (0.35±0.22) with low values (0.08±0.09) during the winter. Summer values are typically higher due to enhanced production of

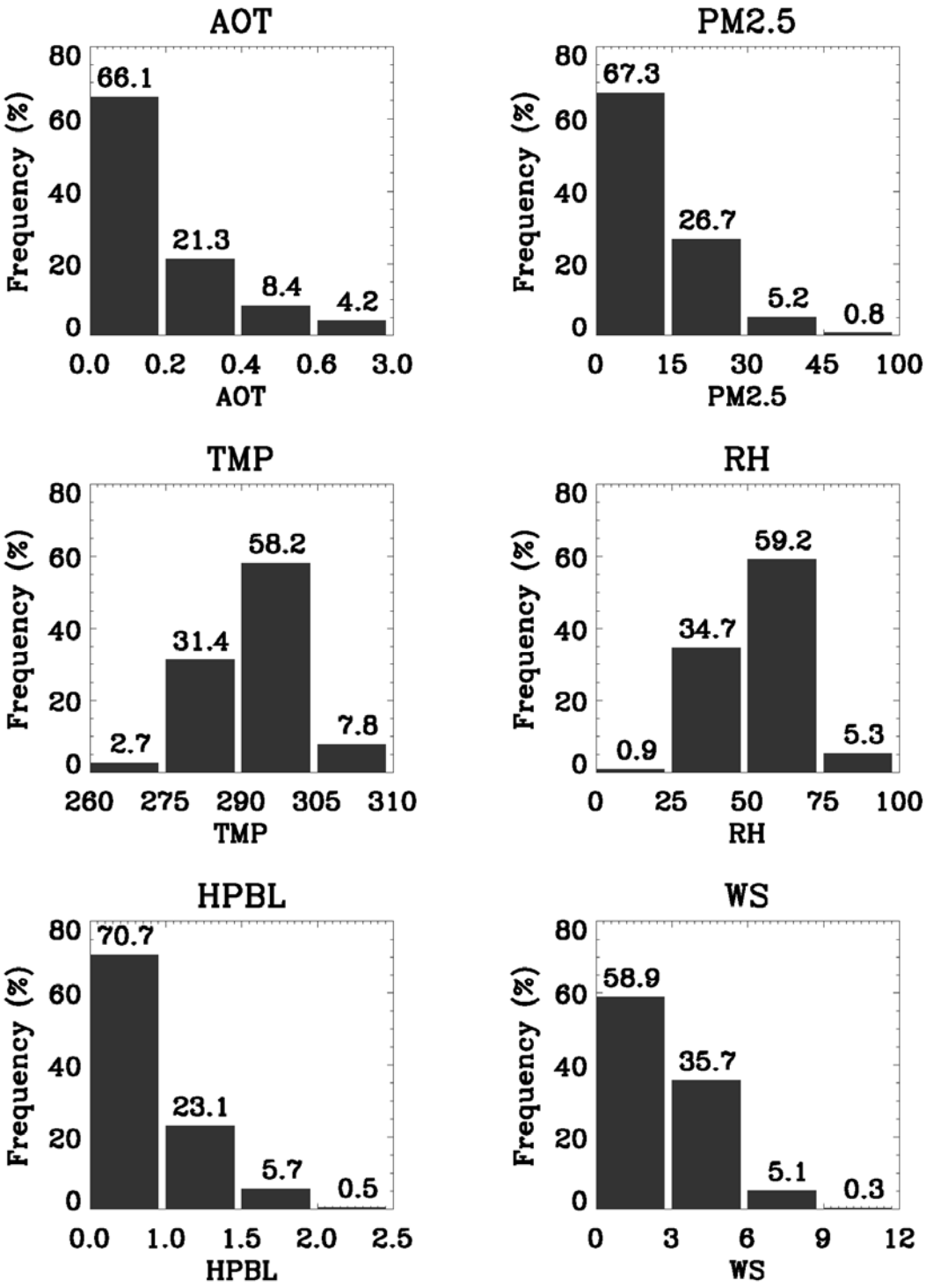

Figure 3.4 Frequency distribution of all the relevant variables used in assessment of particulate matter air quality. AOT is from MODIS satellite, PM2.5 is from ground measurements, and TMP, RH, HPBL and WS are from RUC reanalysis.

secondary particles and enhanced scattering efficiencies of hygroscopic particles. Summer mean AOT values are 40-50% higher when compared to mean AOT values in spring (0.18±0.16) and fall (0.14±0.15). Similar seasonal variations are also seen in PM2.5 mass concentration with largest mean value (20.3±10.5 μgm^{-3}) observed in summer and lowest mean (8.6±6.2 μgm^{-3}) in winter season. Mean PM2.5 mass concentration in fall (13.4±9.4 μgm^{-3}) and spring (11.6±7.2 μgm^{-3}) seasons is almost the same.

Recently, USEPA has revised the standard for its daily averaged PM2.5 mass concentration limit from 65μgm^{-3} to 35μgm^{-3} whereas the annual average standard remains 15μgm^{-3}. The range of PM2.5 mass shows (Figure 3.4) that air quality over these stations reached moderate to unhealthy categories at certain times but their frequencies are much lower (30% for moderate and 2% for unhealthy) when compared to good (68%) air quality conditions. The HPBL have the highest values during summer with mean values of (0.94±0.5 km), which is about 50% higher than HPBL values in winter (0.46±0.29 km). This is to be expected because of higher solar heating during summer seasons. Relative humidity varies between 47% and 60% for different season with highest in summer and lowest in winter. Since the TEOM measures the $PM_{2.5}$ mass under RH conditions between 40-50% the $PM_{2.5}$ values represent dry aerosol mass, and under dry conditions (RH<50%) MODIS AOT is more representative of $PM_{2.5}$ mass concentration.

3.5 Results and Discussions

The results and discussions are organized as follows. Section 5.1 discusses and compares the performance of the two methods (TVM and MVM). Section 5.2 discusses the seasonal differences in the methods. Section 5.3 describes results from MVMs derived for every 2x2 degree sub-region to assess the AOT-PM2.5 relationships due to changes in geographical location. In the last section (5.4), the impact of meteorological parameters on these relationships is analyzed.

3.5.1 Performance of Two Variate and Multi-Variate Methods

We first evaluate the MVM by comparing it with the two variate AOT-PM2.5 method. The linear correlation coefficients (R) and slopes (M) of the regression lines between estimated and observed PM2.5 mass is calculated for each PM2.5 location. Figure 3.5(top panel) shows the change in R value for each station (x-axis). Red and blue lines show the absolute values of R for the TVM and MVM respectively whereas the green line shows the percentage improvement in R value while using MVM when compared to the TVM. The R values between the estimated and observed PM2.5 using the TVM varies from 0.18 to 0.81 with a mean value of 0.60. Positive values of fractional improvement in R (green) clearly shows that inclusion of meteorology in the analysis improved the estimation of PM2.5 mass concentration over each station. The improvement in R value varies for each station and ranges from 5% to almost a three fold increase with an average improvement of 21%. The stepwise regression analysis shows that there is about 7% increase in correlation when surface temperature (TMP) is added to the two-variate model. This improvement has been observed for all other variables as they are added in a stepwise manner to the analysis (Table 3.2).

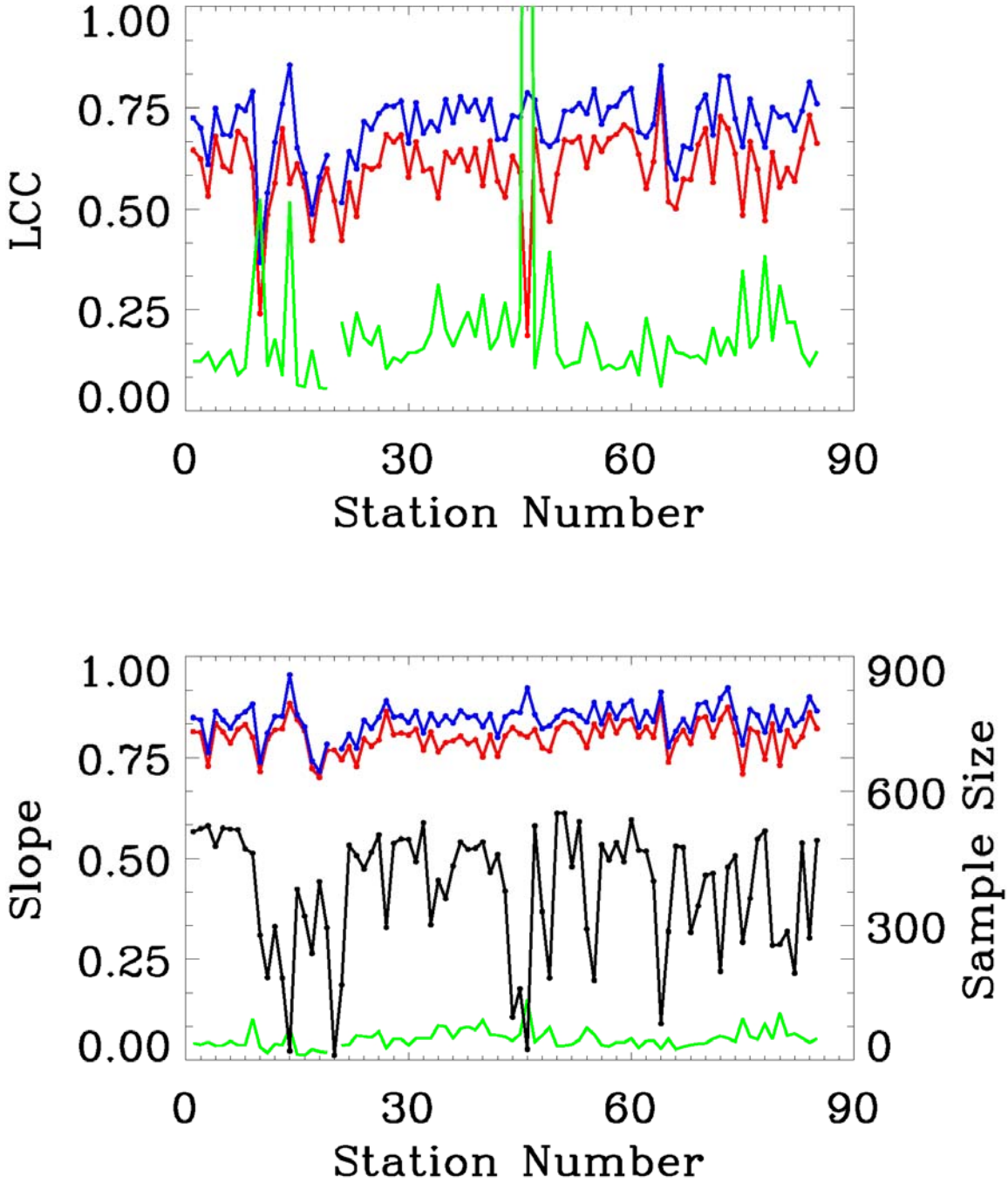

Figure 3.5 Variations in linear correlation coefficient and slope as function of stations for two-variate and multi-variate models. Also plotted the number of data points used in the regression equation on secondary y axis (bottom panel).

Table 3.2 Linear correlation coefficient (R), slope and intercept for different multi-variate models to estimate PM2.5 mass concentration as function of different independent variables.

Model No.	**Independent Variable**	**R**	**Change in R (%)**	**Slope**	**Intercept**
1	AOT	0.604		0.37	8.6
2	AOT, TMP	0.648	7.3	0.42	7.8
3	AOT, TMP, RH	0.662	9.6	0.43	7.6
4	AOT, TMP, RH, HPBL	0.670	10.9	0.45	7.3
5	AOT, TMP, RH, HPBL, WS	0.677	12.1	0.45	7.2
6	AOT, TMP, RH, HPBL, WS, CF	0.683	13.1	0.46	7.1

This 21% improvement in correlation coefficient reduced the error (percentage error of estimation) by 13% and 17% in estimating hourly and daily average PM2.5 mass concentrations respectively.

The MVM estimates PM2.5 mass using satellite and meteorological field with an average error of 34% and 24% for hourly and daily PM2.5 averages. There is some variation in this error over different areas as well as with seasons. Improved estimation of PM2.5 from satellite observations can provide a tool to monitor air qualities in the areas where surface monitoring stations are not available. Also, further improvement and modification to current MVM can serve as a valuable air quality assessment tool in such areas. The MVM linear correlation value ranges from 0.37 to 0.86 with mean value of 0.71. Some locations have very limited sample size (<100) as shown in Figure 3.5(bottom panel). These stations (14, 20, 44, 46, and 64) are shown in Figure 3.2. Due to very small (25) sample size over station number 46 (MS), the improvements in correlation should be interpreted with caution. Similarly, station numbers 14 and 20 also have low sample sizes.

Figure 3.5(bottom panel) shows the slope of the linear relationship between estimated and observed PM2.5 using the TVM and MVM and the corresponding improvements. Slope values have also increased for MVM and the improvement ranges from 1% to 15% with mean value of 5%. The small ranges of slope (0.7 to 0.88 for TVM, and 0.72 to 0.95 for MVM) shows the stable nature of PM2.5-AOT relationships from station to station. However, analysis of the actual data over every single station shows a significant reduction in the scatter between estimated and observed PM2.5 mass when

meteorology is included in the analysis. A slope value close to 1 indicates that the estimated and observed PM2.5 mass concentrations are very close to each other. Slope values obtained from MVM has definitely improved the estimation and has also reduced the error in deriving air quality index.

Figure 3.6 shows some selected examples of the scatter plots between observed and estimated PM2.5 mass using the two methods (Figure 3.2). The four panels on the left side (red color symbol) show the scatter plot between observed and estimated PM2.5 mass using TVM whereas the panels on the right (blue colored symbol) are from the MVM. The bottom two panels (2 stations) show the cases where improvement in estimation is achieved while using MVM whereas the top two panels (2 stations) shows no improvement. Further analysis shows that both the stations (top two, 10 and 18) are located close to the coastal region, where uncertainty in the MODIS retrieval could be high [*Remer et al.*, 2005] and the improvement in estimation is due to inclusion of meteorology. Stepwise multiple regression analysis over these four stations (Figure 3.6) reveals that different meteorological parameters are responsible for the improvement over each site. The first order improvement at station 10 is mainly due to inclusion of HPBL whereas second order improvement is due to inclusion of MODIS cloud fraction (CF). However, note that this is a coastal site and the original relationship between AOT and PM2.5 is not strong due to uncertainties associated with MODIS AOT retrievals. Similar analysis at station number 18 shows that HPBL, CF, and WS equally contributed to the observed slight (0.03) improvement. But, Figure 3.6 (station number 18) clearly shows that underestimation in the TVM at lower range of PM2.5 has improved significantly due to inclusion of meteorological parameters.

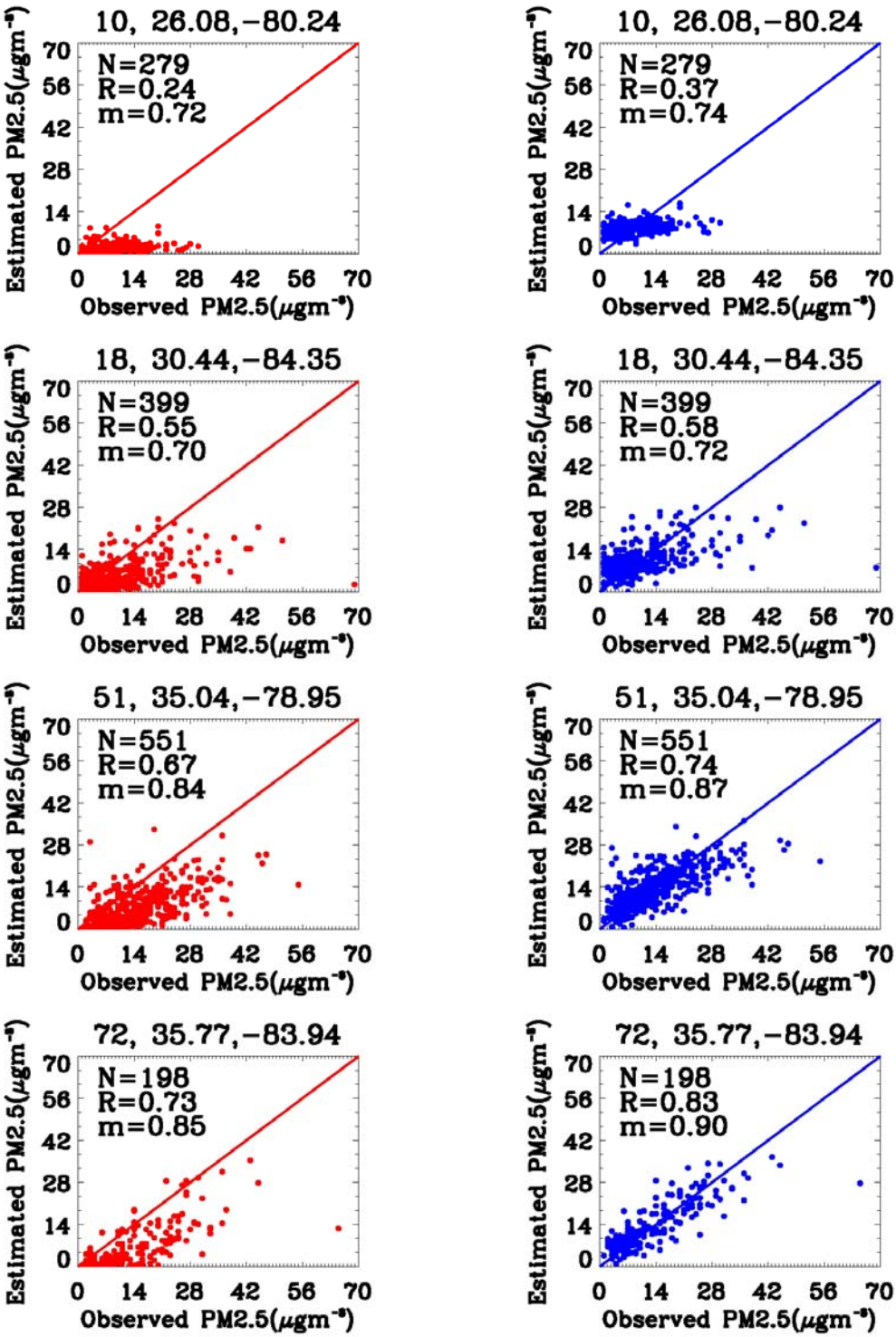

Figure 3.6 Scatter plot for two-variate and multi-variate model estimated PM2.5 and observed PM2.5 for selected stations. Left panels (red) shows results from two- variate model whereas right panels (blue) shows the results from multi-variate model.

A first order improvement at station number 51 and 72 (bottom two panels in the Figure 3.6) is mainly due to inclusion of temperature whereas all other model variables contributed almost equally. Root mean square error (RMSE) values are reduced by 46% (10 to 5.4 μgm^{-3}) and 40% (9.5 to 5.6μgm^{-3}) for station number 72 and 51 respectively when MVM model is used over the TVM model. On the other hand, station number 10 and 18 present only about 20% reduction in RMSE values.

In all the cases, the estimated PM2.5 mass is underestimated by both the methods especially for high (PM2.5>45 μgm^{-3}) PM2.5 mass concentrations. Slope values of <1.0 confirm this underestimation over all stations. There could be many possible causes for this underestimation. One reason is due to inadequate number of data points (<1%) of higher (>45 μgm^{-3}) PM2.5 mass concentration values in the data sets used to form and assess these relationships (Figure 3.4). In most of the cases these high concentration of PM2.5 mass could be associated to specific pollution event or transport of the pollution from different source regions.

3.5.2 Seasonal Analysis

The seasonal analysis is focused on understanding the differences in model slopes and correlation coefficients as well as the change in performance of the two methods over each season. Seasons are defined as spring (MAM), summer (JJA), fall (SON) and winter (DJF). Figure 3.7(a-d) presents the scatter plots between observed and estimated PM2.5 mass concentration using MVM model for each season. Each panel also shows the correlation coefficient ($R_{2,}$ red colored) derived using TVM model. The sample sizes for MAM (8878), SON (8426), JJA (7874) and DJF (7278) are approximately the same.

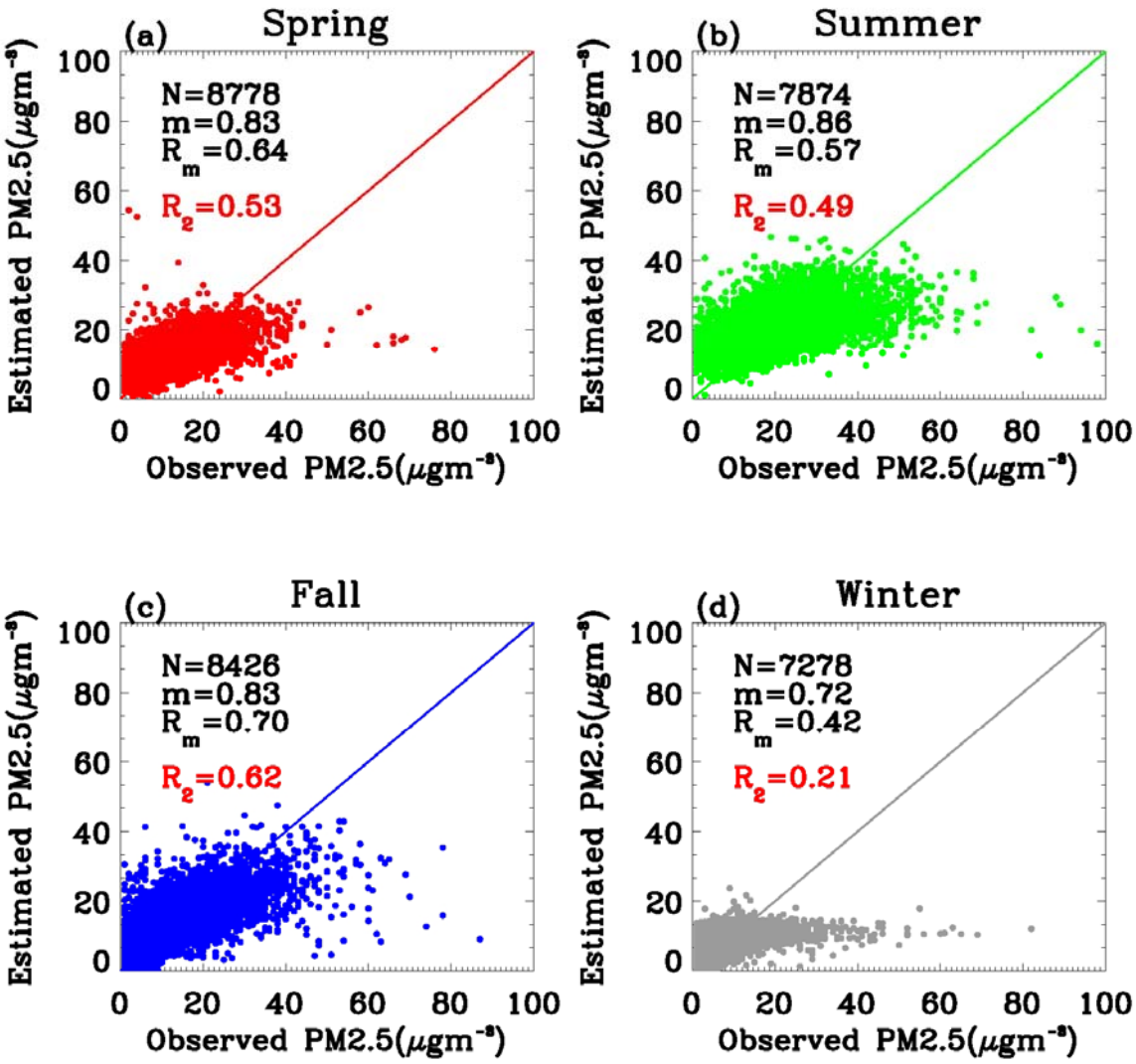

Figure 3.7 Four panel seasonal scatter plots showing observed and estimated PM2.5 using multi-variate regression model as function of seasons. Here DJF is December-January-February, MAM is March-April-May, JJA is June-July-August and SON is September-October-November.

Slope values are less than one for all four seasons thereby leading to an underestimation in PM2.5 mass by both the methods. Again this underestimation in PM2.5 is mainly due to low number of samples of high PM2.5 values in the analysis. Other meteorological parameters such as vertical temperature gradient, emission factors, removal processes may also be responsible, which controls the particle concentration at the surface and have not been included in current analysis. Obviously, accurate estimation of PM2.5 from these methods require accurate input parameters and any uncertainty in input variables will propagate to the output and will have affects on estimated air quality index. The estimations are the best during spring and fall seasons that have the highest number of data points. The methods perform better during summer whereas it is very poor in winter season, although MVM shows improvement in correlation from 0.21 to 0.42 compared to TVM during winter. Percentage change in error of estimation is highest during fall (13%) and spring (13%) whereas it is least in summer (6%) with moderate (10%) in winter season. In all four cases, high values of PM2.5 mass concentration (>45 μgm^{-3}) were underestimated by the model compared to low values of PM2.5.

The poor performance during the winter season is largely due to low AOT values, which are associated with high uncertainties [*Remer et al.,* 2005]. This conclusion is drawn on the basis of very low correlation values in TVM model results as shown R_2 in each panel of Figure 3.7. Similar results were observed in a study over Sydney, Australia where AOT values were less than 0.1 [*Gupta et al.,* 2008]. Including meteorology in the analysis (i.e. MVM model) improves the results. Again HPBLs are observed to be lower in winter by almost 50% than during the summer season. Lower HPBLs in the winter season that do not allow mixing of pollution in longer air column plays an important role

in underestimation of PM2.5 during this season. Section 5.2 discusses more details on the impact of meteorology, which varies between seasons that leads to seasonal differences in estimation of PM2.5 mass.

3.5.3 Geographical Differences

Geographical differences in model coefficients are analyzed by assessing the methods for every 2 degree latitude by 2 degree longitudes over the South Eastern United States. Data from all the stations falling in each grid are grouped and the TVM and MVM as function of seasons are developed. Figure 3.8 shows the scatter plots between observed and estimated PM2.5 mass using MVM models for every grid box. The numbers shown in red inside each scatter plot shows the R values for the TVM model and are used to compare the results from MVM model. The grid between 31-33N and 90-92W did not have any data during winter and spring season over the entire three year time period. A Comparison with Figure 3.2 shows that this grid box contains only one station (42), which has limited number of observations (39 in the summer and 43 in the fall). Again the seasonal pattern over each grid shows almost similar pattern with small variations as seen earlier. The number of available data points and model performance varies with their geographical locations. Correlation coefficients shows good improvement for each box and the degree of improvement varies as a function of location and season. Table 3.3 summarizes minimum, maximum, mean and standard deviation of improvements in correlation and percentage error of estimation for all the data and for each season. Maximum improvement in mean absolute percentage error of estimation (APE) is observed during fall (19%) and winter (18%) whereas it is 15% and 11 % during spring and summer whereas it the reduction ranges from 1% to 45% over different grid box.

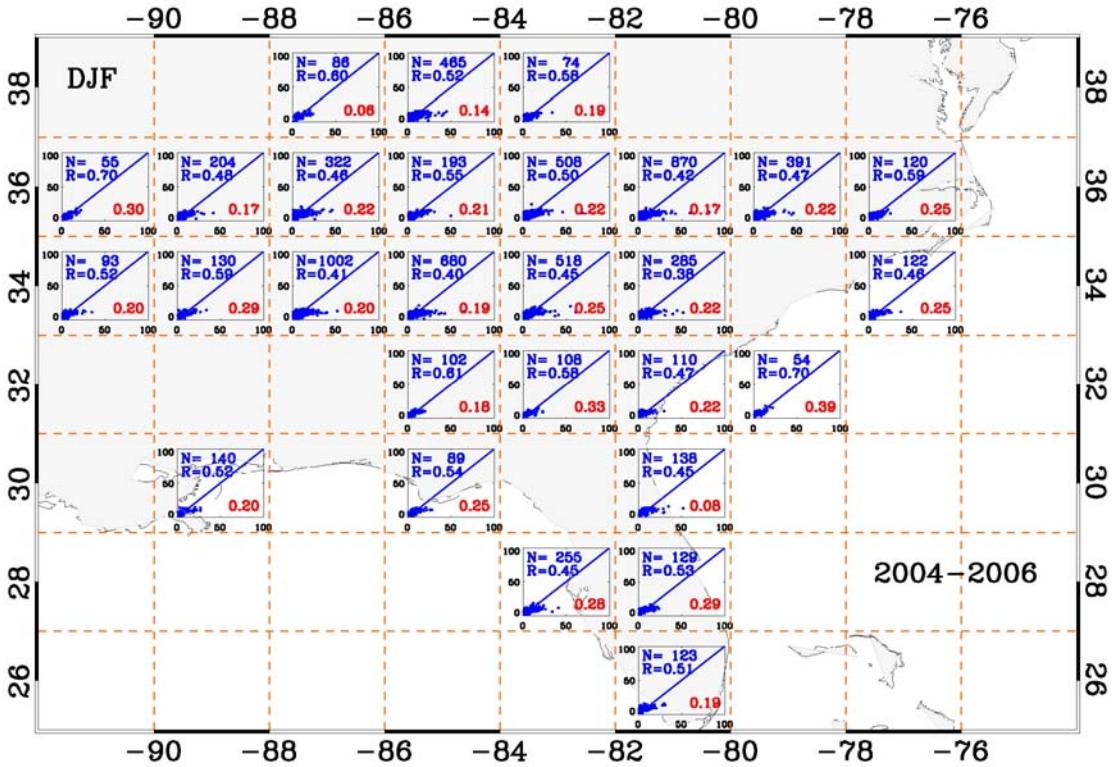

Figure 3.8a Seasonal (winter) map showing scatter plot between estimated and observed PM2.5 in each two by two degree grid box using multi-variate models. Also presented is linear correlation coefficient value (red) for two-variate model.

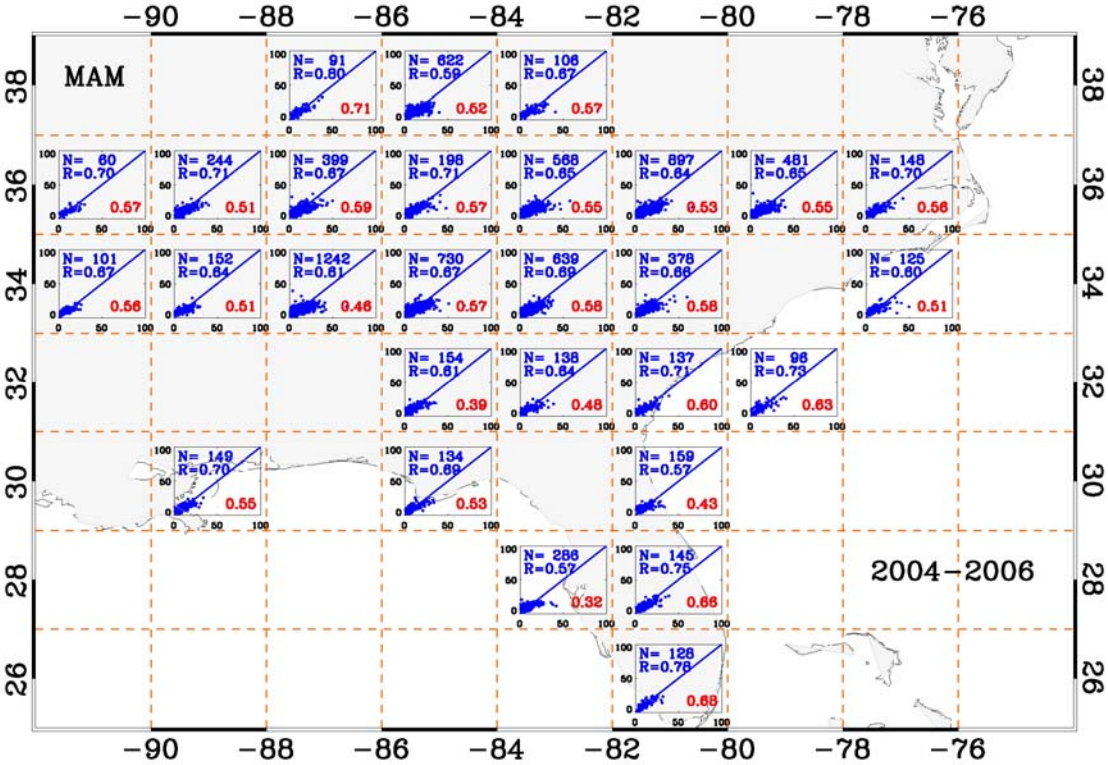

Figure 3.8b Seasonal (spring) map showing scatter plot between estimated and observed PM2.5 in each two by two degree grid box using multi-variate models. Also presented is linear correlation coefficient value (red) for two-variate model.

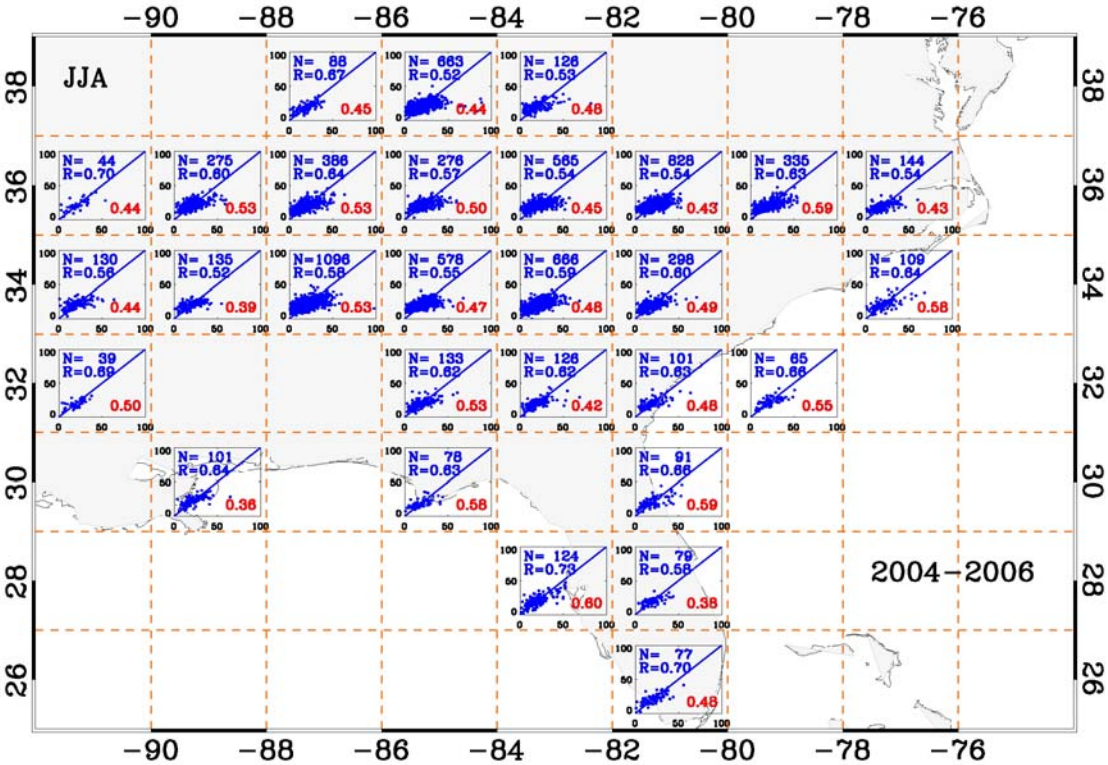

Figure 3.8c Seasonal (summer) map showing scatter plot between estimated and observed PM2.5 in each two by two degree grid box using multi-variate models. Also presented is linear correlation coefficient value (red) for two-variate model.

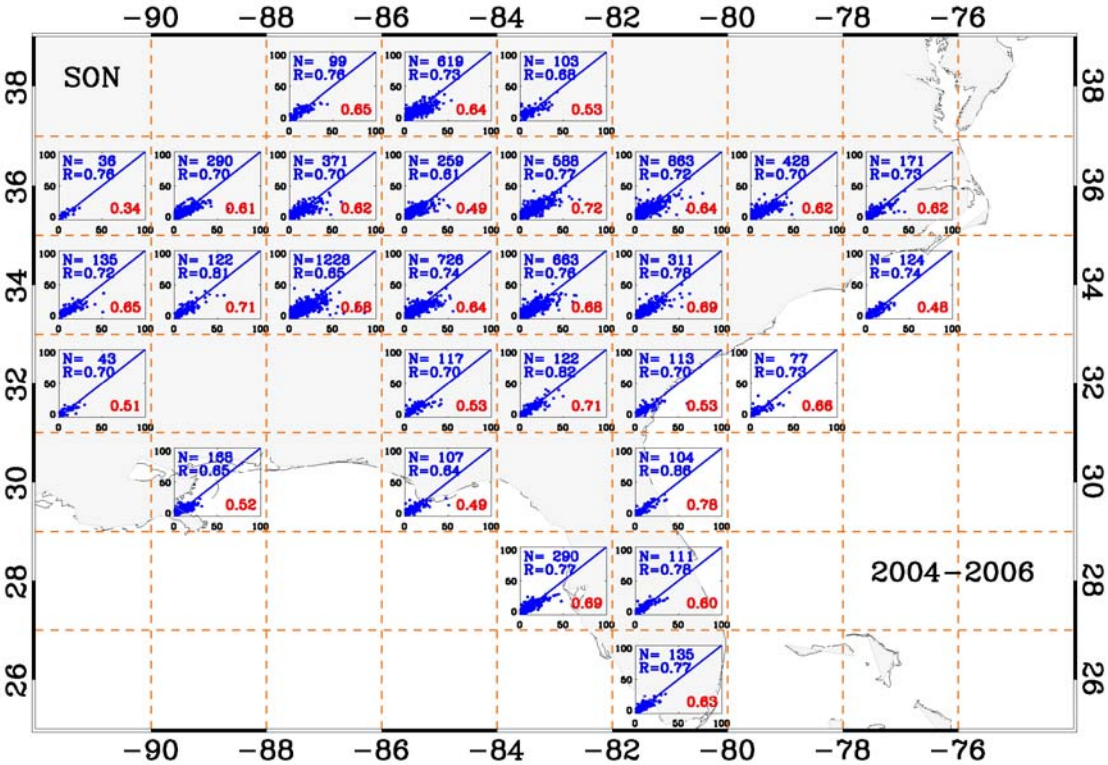

Figure 3.8d Seasonal (fall) map showing scatter plot between estimated and observed PM2.5 in each two by two degree grid box using multi-variate models. Also presented is linear correlation coefficient value (red) for two-variate model.

Table 3.3 Summarized statistics from geographical two-variate and multi-variate model performance as a function of seasons. Absolute percentage error (APE) and correlation coefficient (R) is presented for two-variate and multi-variate models and percentage improvements.

Season	**Parameter**	**TVM-APE (%)**	**MVM-APE (%)**	**AEE-IMP (%)**	**TVM-R**	**MVM-R**	**R-IMP (%)**
Winter	Minimum	42	32	5	0.06	0.38	37
	Maximum	54	48	42	0.39	0.70	91
	Mean	49	41	18	0.21	0.51	57
	Standard Deviation	3	5	10	0.08	0.09	14
Spring	Minimum	33	27	8	0.32	0.57	11
	Maximum	44	37	24	0.71	0.80	43
	Mean	38	33	15	0.55	0.67	19
	Standard Deviation	2	2	3	0.08	0.06	7
Summer	Minimum	30	25	1	0.36	0.52	7
	Maximum	39	39	38	0.60	0.73	44
	Mean	33	30	11	0.49	0.61	20
	Standard Deviation	2	3	8	0.06	0.06	10
Fall	Minimum	36	28	11	0.34	0.61	7
	Maximum	47	38	45	0.78	0.86	55
	Mean	41	35	19	0.61	0.73	17
	Standard Deviation	3	3	7	0.09	0.06	10

3.5.4 Impact of Meteorology

The impact of meteorology on model performance is evaluated by analyzing the model results as function of each meteorological parameter. These include HPBL, TMP and RH. Figures 3.9, 3.10 and 3.11 show the performance of the MVM as a function of HPBL, RH and TMP respectively. Each panel in these figures also shows the number of data points (N), linear regression line (Y=mX), linear correlation coefficients (R_m: for MVM and R_2: for TVM). Multi variate models are developed for four different ranges of HPBL; surface to 1km, 1 to 1.5 km, 1.5 to 2.0 km and 2 to 2.5 km (Figure 3.9). Note the number of available data points in each bins reduced drastically for higher HPBL values, which shows that region of study does not experience frequent high mixing layer during the satellite (Terra) overpass time. The HPBL significantly impacts the relationship between PM2.5 and AOT. Linear correlation coefficient values derived for TVM (R_2) in each PBL bin shows significant improvement under high PBL conditions and varies between 0.57 and 0.73 for low to high HPBL bins. Similarly, correlation values for MVM model also shows improvement for higher HPBL bins. Correlation is highest (0.8) between estimated and observed PM2.5 values for HPBL > 2 km. The higher correlation for higher HPBL clearly shows that, under a well mixed boundary layer, satellite measurements (and hence retrievals) of aerosols represents surface level PM2.5 more accurately.

Figure 3.10 presents the scatter plots between observed and estimated PM2.5 mass using MVM for four different bins of RH. The model performance is best for RH range of 25% to 50%, which is also close to PM2.5 measurement condition (40%) in

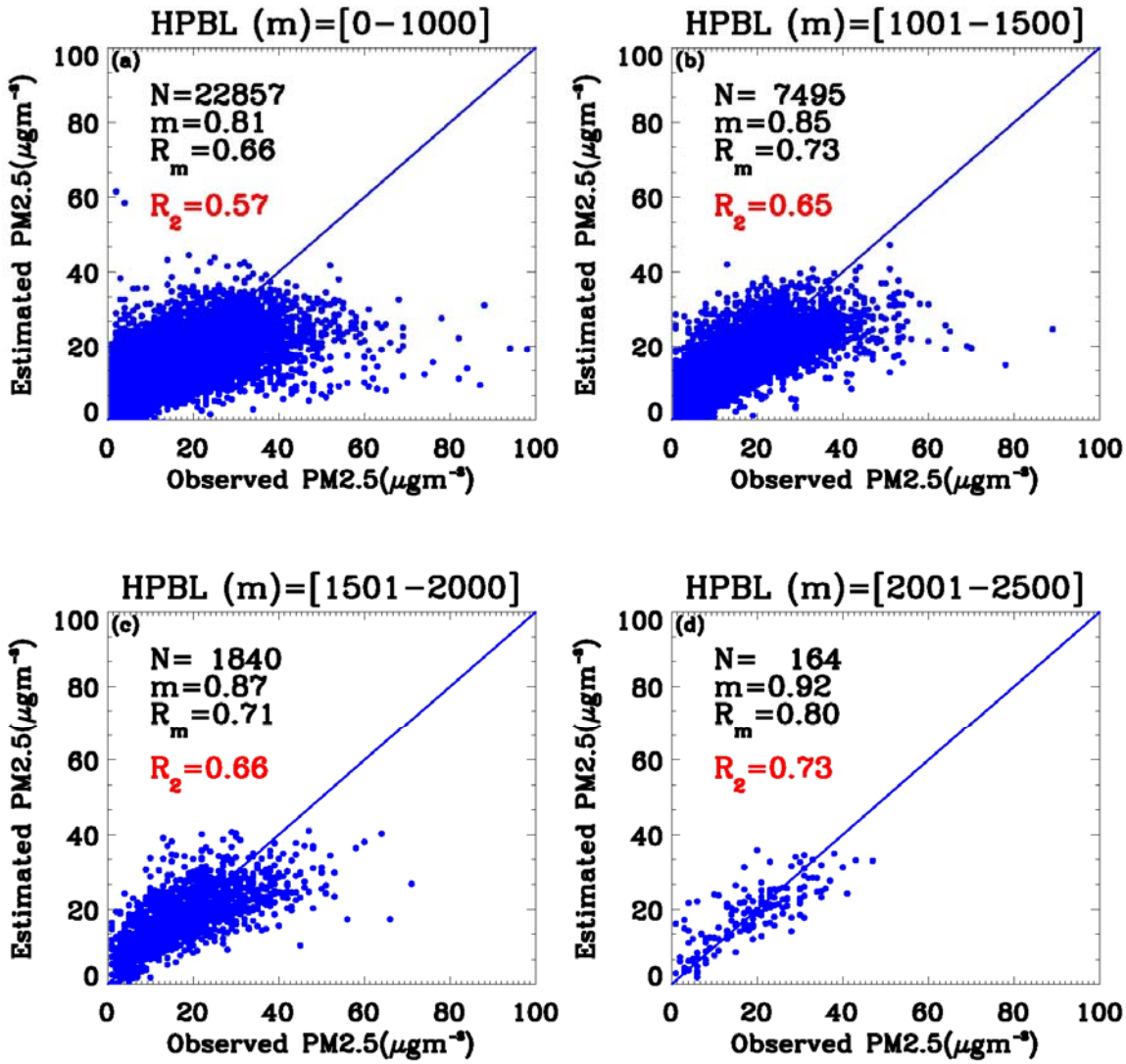

Figure 3.9 Scatter plot between observed and estimated PM2.5 mass concentration using multi-variate model for four different bins of height of planetary boundary layer.

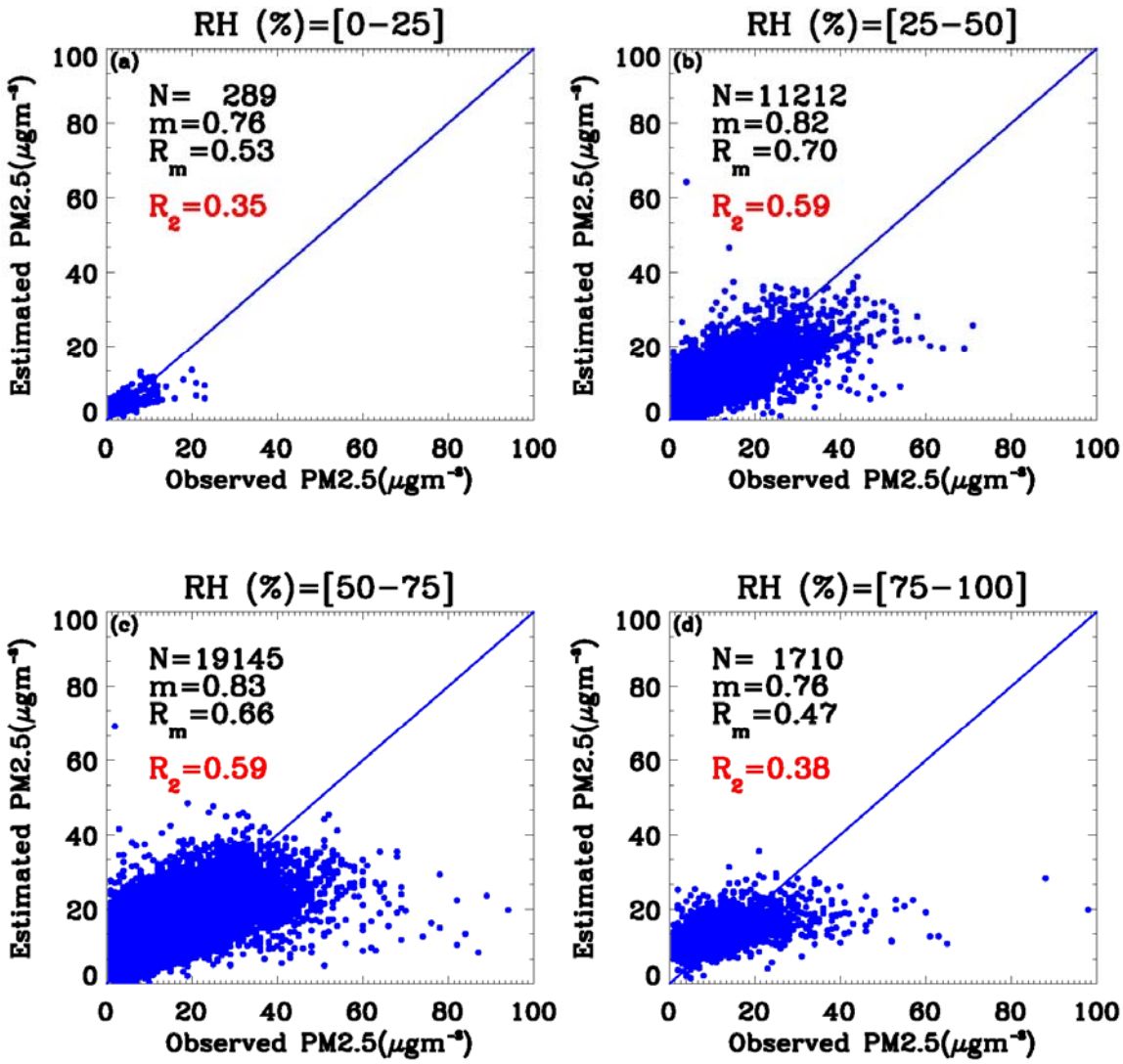

Figure 3.10 Scatter plot between observed and estimated PM2.5 mass concentration using multi-variate model for four different bins of relative humidity.

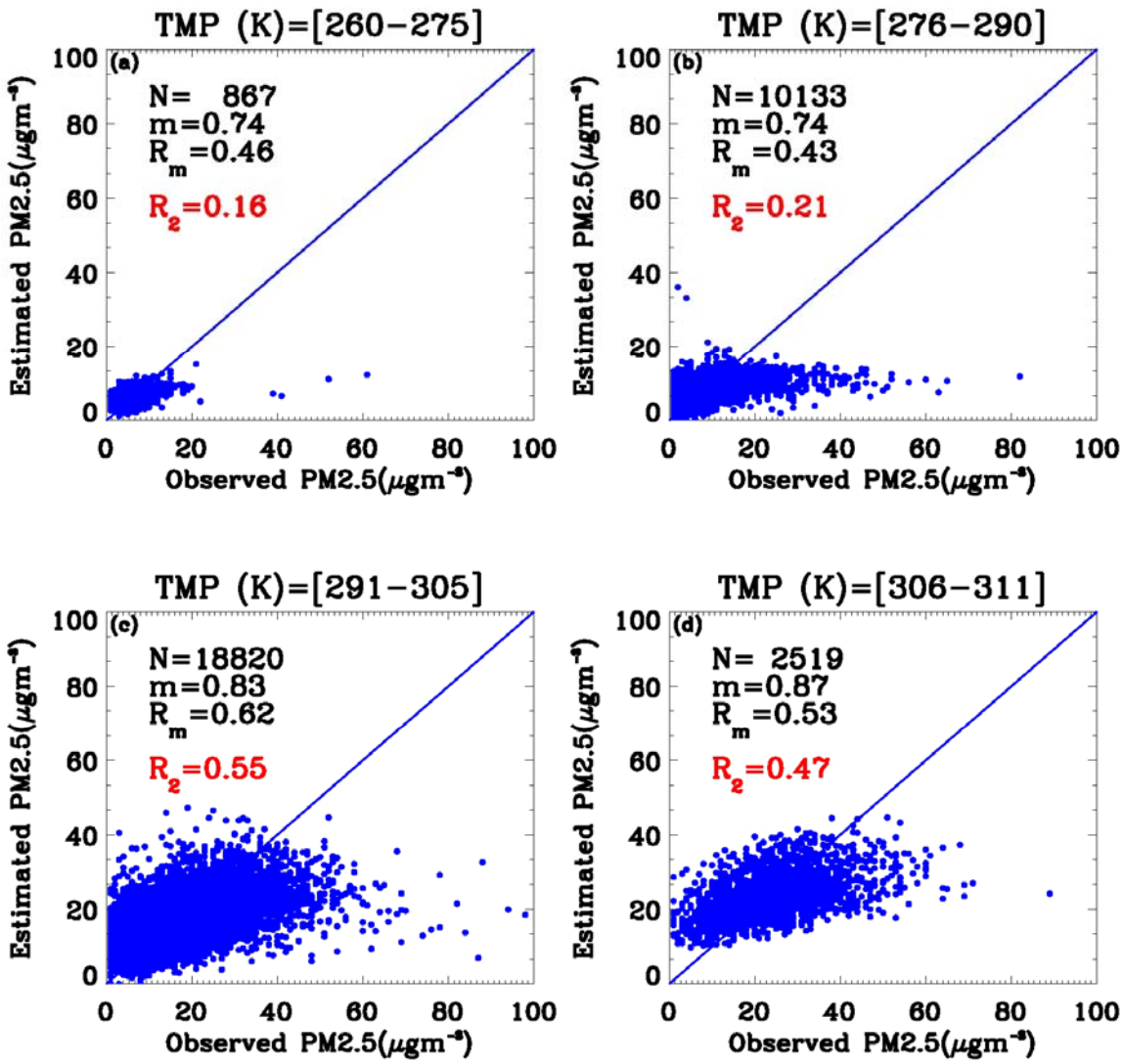

Figure 3.11 Scatter plot between observed and estimated PM2.5 mass concentration using -variate model for four different bins of surface temperature.

TEOM. Also, it is important to note that under all ranges of humidity conditions; the estimation of PM2.5 is improved using MVM when compared to the TVM. Similarly, Figure 3.11 shows the impact of surface temperature on model performance. TMP also impact the model performance but it is relatively small and variations in model parameters are less for different range of TMP. High temperature conditions can enhance the emission of particles from surface as well as accelerate the formation of secondary particles in the atmosphere.

3.6 Summary and Conclusions

Satellite remote sensing is an emerging and promising tool for monitoring aerosol pollution at global scales with high temporal and spatial resolution [*Al-Saadi et al.,* 2005*; Fishman et al.,* 2008]. This is especially attractive in regions or countries where ground measurements are not available or not possible and remote sensing due to its reliability can serve as a cost effective tool. However satellites provide columnar measurements and ancillary pieces of information is required to convert this columnar measurement to surface values. A commonly used method is to form linear relationships between columnar satellite-derived AOT and surface PM2.5 from ground monitors and to extend these relationships to other regions where ground monitors are not available. Aerosol observations from both geostationary and polar orbiting satellites have been used successfully to assess this method. In this study, we take this analysis a step further to see if meteorological fields can help improve this AOT-PM2.5 relationship.

Therefore, aerosol measurements from MODIS-Terra, meteorological parameters from RUC20, and PM2.5 mass concentrations from surface stations over entire southeast

United States for a three year time period have been analyzed for assessing surface level PM2.5. Multi variable statistical models have been developed as function of regions and season to estimate surface level PM2.5 mass concentration while making use of satellite and model data sets. Our results indicate that multi variable models provide improved estimations of PM2.5 mass concentration over two variable regression equations used in previous studies [*Wang and Christopher et al.,* 2003; *Engel-Cox et al.,* 2004]. Linear correlation coefficients between observed and estimated PM2.5 mass concentration show an average improvement of 21% over different stations in the region while using the MVM model over TVM model. Specifically, improvements are seen during spring and fall compared to summer season while the results during winter are very poor. The seasonal differences in performance of model are mainly associated with satellite data quality, type and level of pollution and the highly variable nature of local meteorological conditions which plays an important role in formation and removal of PM2.5 in the earth atmosphere. Geographical differences in accuracies of estimated PM2.5 mass are observed when models were developed for each 2 degree by 2 degree region.

In part 2 of this series, we plan to extend the idea of using meteorological fields to estimate PM2.5 mass concentration from satellite remote sensing data in an artificial intelligence frame work. The significant conclusions from this study are as follows:

1. The MVM provides better estimations of surface PM2.5 mass concentration when compared to TVM and it is a function of location and season.
2. Stepwise multiple regression analysis shows that first order influence on PM2.5-AOT relationship is produced by temperature when included in the

regression and second order impact is due to inclusion of boundary layer height. It is also important to note that the order of importance changes with season and location.

3. The MVM provides improved estimation (R>0.8) of PM2.5 mass concentration under high (>2 km) planetary boundary layer conditions due to well mixed nature of aerosol layer.
4. There are definite seasonal differences in model performance. Models perform best during fall (R=0.70), average during spring (R=0.64) and summer (R=0.57) and below average during winter (R=0.42) season.
5. Overall, both models tend to underestimate PM2.5 mass concentration during heavy pollution days under all meteorological conditions.

Although the application of satellite observations in monitoring particulate air quality is useful, there are many issues in applying satellite derived AOT to estimate surface level PM2.5 mass concentration. Most researchers in this field agree that AOT alone can not be used to estimate surface level pollution with good accuracies. Theoretically, surface level PM2.5 is influenced by various meteorological parameters and therefore including information on local meteorology should improve the estimation accuracies. The current study has used a large data base from satellite and surface to demonstrate that there is significant reduction in the error (13%) when local meteorology is used in conjunction with satellite data. The current approach shows that without using any complex physical and numerical models, simple statistical models can be used to estimate PM2.5 with an average uncertainty of 34% for hourly and 24% for daily mean mass concentrations.

CHAPTER 4

AIR QUALITY ASSESSMENT USING ARTIFICIAL NEURAL NETWORK

4.1 Introduction

Satellite remote sensing of fine particles (PM2.5, aerodynamic diameter less than 2.5 μm) is an innovative and relatively new technique that is used to assess surface level PM2.5 mass concentration with high spatial and temporal resolution. A study by *Wang and Christopher* [2003] first showed that under certain conditions, PM2.5 mass concentration measured at the surface and aerosol optical thickness (AOT) from MODIS-Terra and MODIS-Aqua satellites are well correlated (R>0.7). They also reported that AOT measurements can be used to estimate PM2.5 mass concentration. Other studies [*Fraser et al.,* 1984; *and Sifakis et al.,* 1998*; Chu et al.,* 2003] have also reported the potential of satellite measurements for PM2.5 air quality monitoring in areas where surface measurements were not available. Study by *Fraser et al.,* [1984] estimated the columnar sulfate concentration (gm^{-2}) over a few locations on east coast of United States using AOT measurements from the Visible Infrared Spin-Scan Radiometer (VISSR) onboard Geostationary Operational Environmental Satellite (GOES).

Similarly, a good degree of correlation (0.76) was found between Landsat TM derived AOT and surface measured SO_2 mass concentration in Athens [*Sifakis et al.,* 1998].

Since the study by *Wang and Christopher*, [2003], several papers have been published that have utilized satellite-derived AOT as a surrogate for PM2.5 mass concentrations (Table 4.1). Table 4.1 shows that most of these studies [*e.g. Engel-Cox et al.,* 2006; *Hutchinson et al.,* 2006; *Al-Saadi et al.,* 2005] were largely focused on the United States. They also used MODIS satellite data to estimate surface level PM2.5 mass concentration. MODIS was designed specifically for aerosol studies with good calibration and state of the art retrieval algorithms to convert measured radiances to AOT values. 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 satellite also provides reliable AOT retrievals [*Diner et al.,* 2000] and this data has been also used to characterize PM2.5 mass concentrations over the United States [*e.g., Liu et al.,* 2008]. However, due to its narrow swath width, MISR global coverage is only achieved on a weekly basis. Recently, AOT data from geostationary satellites (GOES) have been also evaluated for PM2.5 mass estimation at higher temporal resolutions [*Paciorek et al.,* 2008].

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 activities. Most of these studies are based on correlating AOT and PM2.5 with simple linear regressions. The MODIS derived AOT which is measure of column

Table 4.1 Literature survey on satellite remote sensing of particulate matter air quality from past half decade of research.

S.N.	Reference	Data & 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	*Hutchinson, 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 entire USA and discuss difference in relationship over different regions. Qualitative and qualitative analysis over larger area, demonstrated spatial distribution of correlation. Range of R.
5	*Hutchinson et al., 2004*	MODIS AOT maps, Ozone, Eastern USA	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, USA	First used MISR data for air quality study and have emphasis on seasonal and annual mean correlation analysis and forecasting. (R=0.78)
7	*Engel-Cox et al., 2004*	MODIS	Recommendations to use satellite data into air quality applications. These data sets can add synoptic and geospatial information to ground-based air quality data and modeling.
8	*Liu et al., 2005*	MISR , GEOS-3 Meteorology, USA	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.
9	*Al-Saadi, et al., 2005*	MODIS, USA	More descriptive paper on IDEA program, which provides online air quality conditions from MODIS and surface measurements

			over several locations in the USA
10	*Hutchinson 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
11	*Engel-Cox et al., 2005*	MODIS, USA	Potential of satellite data for monitoring transport of PM2.5 over state boundaries and event specific analysis.
12	*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.
13	*Engel-Cox et al., 2006*	MODIS, LIDAR, USA	Weak correlation can be significantly improved by using vertical aerosol information from LIDAR measurements.
14	*van Donkelaar et al., 2006*	MODIS, MISR, PM2.5, GEOS-CHEM, USA and Global	Inter-comparison between MODIS and MISR over several locations in Canada and USA. R= 0.69 (MODIS) and R= 0.58 (MISR). Different approach used to calculate the fine mass concentration.
15	*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.
16	*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.
17	*Liu et al., 2006*	MODIS, MISR, RUC	Inter-comparison between MODIS and MISR in St. Louis area. MISR performed slightly better than MODIS in the region.
18	*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.
19	*Gupta and Christopher, 2008a*	MODIS, Birmingham, AL, USA	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.
20	*Kumar et al., 2007*	MODIS (5km), Delhi, India	Three months PM2.5 data from a field campaign were used. Correlation between

			PM2.5-AOT was 0.52±0.20
21	*Kumar et al., 2008*	MODIS (5km), Delhi, India	Same as Kumar et al., 2007, plus more analysis on PM10-AOD relationships
22	*Gupta and Christopher, 2008b*	MODIS, Southeast USA	Long terms 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 SE USA.
23	*Schaap 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.
24	*Liu et al., 2008a*	MISR, USA	Describe method of estimating PM2.5 mass and its major constituents using fractional AOD values from different aerosol types in MISR algorithm.
25	*Liu et al., 2008b*	MISR, USA	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.
26	*Martin and Canada, 2008*	Review	Mostly focused on gaseous air quality but also provide some review on particulate matter air quality from satellite observations.
27	*Hutchinson et al., 2007*	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.
28	*Paciorek et al., 2008*	GASP, MODIS, MISR, USA	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

aerosol loading 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 (TMP), relative humidity (RH), wind speed (WS) and direction (WD), variations in sunlight due to clouds and seasons are important parameters which affect the relationship between the two parameters. Although AOT-PM2.5 relationships work well in some regions, a major issue is the lack of vertical information since AOT is a columnar quantity whereas the PM2.5 is a surface measurement. Although ground and space-borne lidars are a good solution for obtaining this vertical information, they are not readily available and therefore using meteorological information such as mixing layer heights could be a viable solution.

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 [*Mathur,* 2008]. Monitoring and forecasting of daily air quality based on PM2.5 mass using different forecasting models is still in its infancy [*Kondragunta et al.,* 2008]. Uncertainties exist because the sources of pollutants are not well defined and gaps in our knowledge of physical, dynamical and chemical processes in the atmosphere. New approaches and systematic modeling therefore are highly needed to estimate the air quality in the United States and around the world, especially in areas that experience poor air quality with limited or no ground measurements.

Satellite data have used to form regression models [part 1 of this series, here after referred as *Gupta et al.,* 2008c], 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.,* 1996*; Hubbard et al.,* 1998*; Ryan,* 1994]. To explore the possibility of improvement on these issues, an Artificial Neural Network (ANN) based approach along with satellite, ground and meteorological data sets to monitor air quality is developed. In this paper we compare the results of the ANN method with the two variate and multi-variate methods.

4.2 Artificial Neural Networks

The complexity of a problem and its understanding decides what type of modeling system would be required. A full numerical model would be most suitable for estimation of particulate matter air quality if we have the required data sets and a good understanding of PM2.5 formation and removal processes. But, 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 work, such as the brain process information [*Bishop,* 1995]. In other words ANN is a set of computer algorithms designed to simulate biological neural network in terms of learning and pattern recognition. Artificial neural network has been used by many scientific disciplines to identify patterns and extract trends in imprecise and complicated non-linear data.

Artificial neural network for addressing earth-science problems is not new. For example, ANN has been used to develop cloud detection algorithms for the Polar Regions [*Lee et al.,* 1990]. In recent years ANN have 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.,* 2004*;*

Jiang et al., 2004*; Perez and Reyes,* 2006*; Chattopadhyay and Bandyopadhyay,* 2006]. A study by *Gardner* [1998] in London used ANN to successfully demonstrate the prediction of NO_x and NO_2 by providing inputs such as meteorological condition and traffic flow data. Daily average PM2.5 mass concentration was forecasted in El Paso (USA) and Ciudad Juarez (Mexico) using three different types of neural networks, and two classical models (persistence and linear regression). Results indicated that neural network models outperformed the classical models [*Ordieres et al.,* 2004]. A similar study by *Perez et al.,* [2000] compared neural network based models and classical models to forecast hourly PM2.5 mass concentration using prior day observations in Santiago, Chile. Results from this study again confirm that neural network perform extremely well. These studies were able to conclude that neural network based modeling system perform more efficiently when compared to linear regression models for particulate matter air pollution monitoring and forecasting [*Perez and Reyes,* 2006].

In this paper, several neural network based models (or networks) have been developed to estimate surface level PM2.5 using satellite and meteorological fields as function of season and regions over the Southeastern United States. In our previous study, we have used two variate (TVM) and multi-variate methods (MVM) to estimate the surface level PM2.5 mass concentration. Results show improvement in PM2.5 estimation accuracy (13% and 17% for hourly and daily average respectively) while using meteorology along with satellite observations in MVM models. To our knowledge this is the first time that aerosol observations from satellite and surface meteorology have been used to train an ANN system to estimated hourly and daily average PM2.5 mass concentrations.

In the past, many studies have shown the application of multi layer perceptron (MLP) type of neural network to model air quality and other atmospheric problems. Multi layer 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 patter reorganization, classification) rather than memorizing the training data. For example training data in air quality applications usually comprise of local meteorological conditions, climatological value of pollution level, space and time information, and all those parameters which can determines the pollution level. A trained neural network system behaves in a more generalized manner. This is one of the important advantages over using 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 [*Schakoff,* 1992] that employ regression models.

Common neural networks architecture have three layers of neurons: input layer, hidden layer and output layer. Each of these layers can have one of more than one nodes or neurons. Figure 4.1 provides a schematic of such a network used in the current study with 8 nodes (i.e. input parameters) in input layer and two nodes (i.e. PM2.5 for hourly and 24hour average) in the output layer. The input layer consist of 8 nodes namely latitude, longitude, month, AOT, WS, RH, HPBL and TMP. The input layers are connected to hidden and output layers by linear combination of functions. Layers between input layer and output layer are usually called hidden layers and work towards minimizing the error by modifying weights through training process. Nodes or neurons of a neural network are connected by output signal and weights, which are modified by a

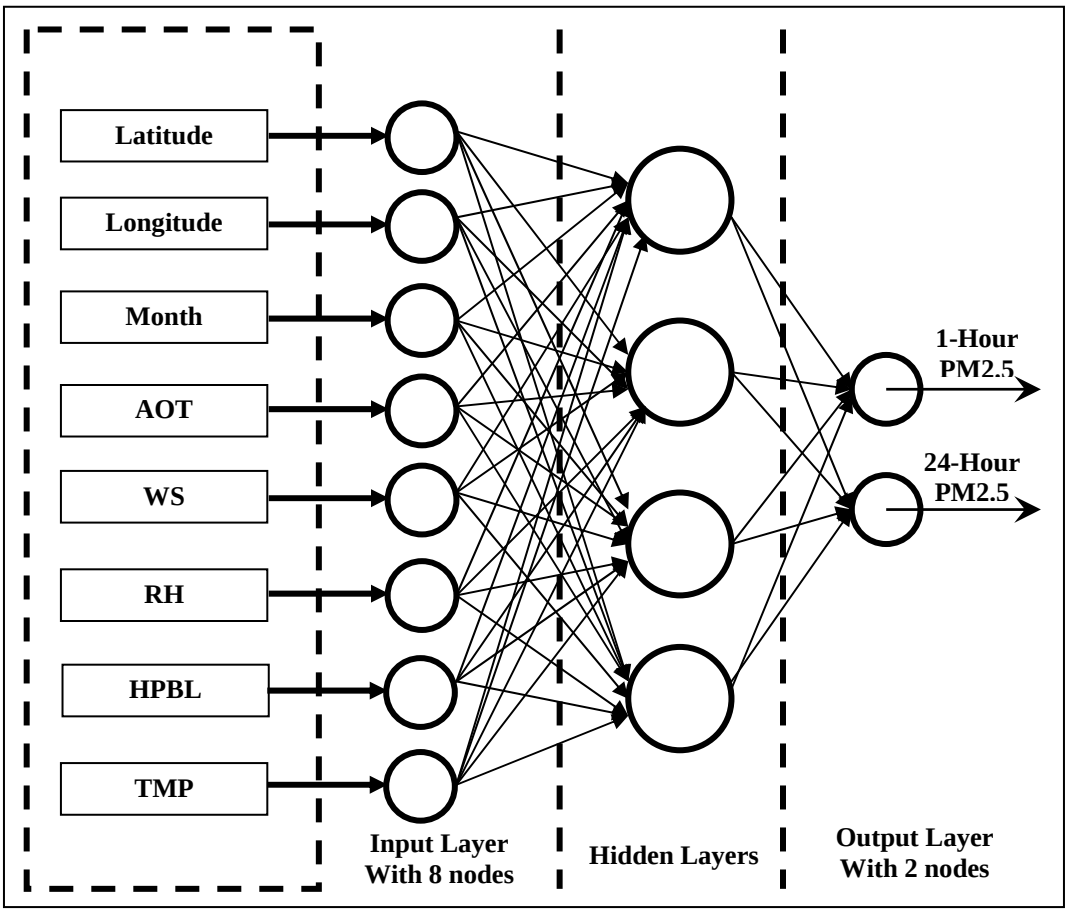

Figure 4.1 Schematic of a multi layer perceptron neural network used to integrate satellite and meteorological fields to estimate surface level PM2.5 mass concentration.

simple nonlinear transfer or activation function [*Gardner and Dorling,* 1998]. Commonly used nonlinear functions among others are logistic, exponential, hyperbolic tangent (tanh) and identity. 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 neural network. During each pass of data, neural network calculates the output (in this case PM2.5 mass concentration), which is compared with desired output (actual or observed PM2.5 measurement) and an error is estimated. This error is then sent back to the network, which force the network to adjust its weight such that the error decreases with each iteration until to the desired outcome is achieved. Hence, training is 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 new input vector. Figure 4.2 provides a flow chart of typical back-propagation training algorithm. A step by step description of training process can be found in [*Gardner and Dorling,* 1998], and further theoretical details can be found in *Bishop* [1995].

4.3 Data Sets and Network Training

The data sets are discussed in detailed in *Gupta et al.,* [2008c] and only a brief description is given here for sake of completeness. Three years of hourly PM2.5 mass concentration (μgm^{-3}) from EPA AirNow ground based air quality network, hourly meteorological fields from Rapid Update Cycle (RUC) reanalysis at 20x20 km^2 spatial resolution, and instantaneous observations of aerosol optical thickness at 0.55 µm from

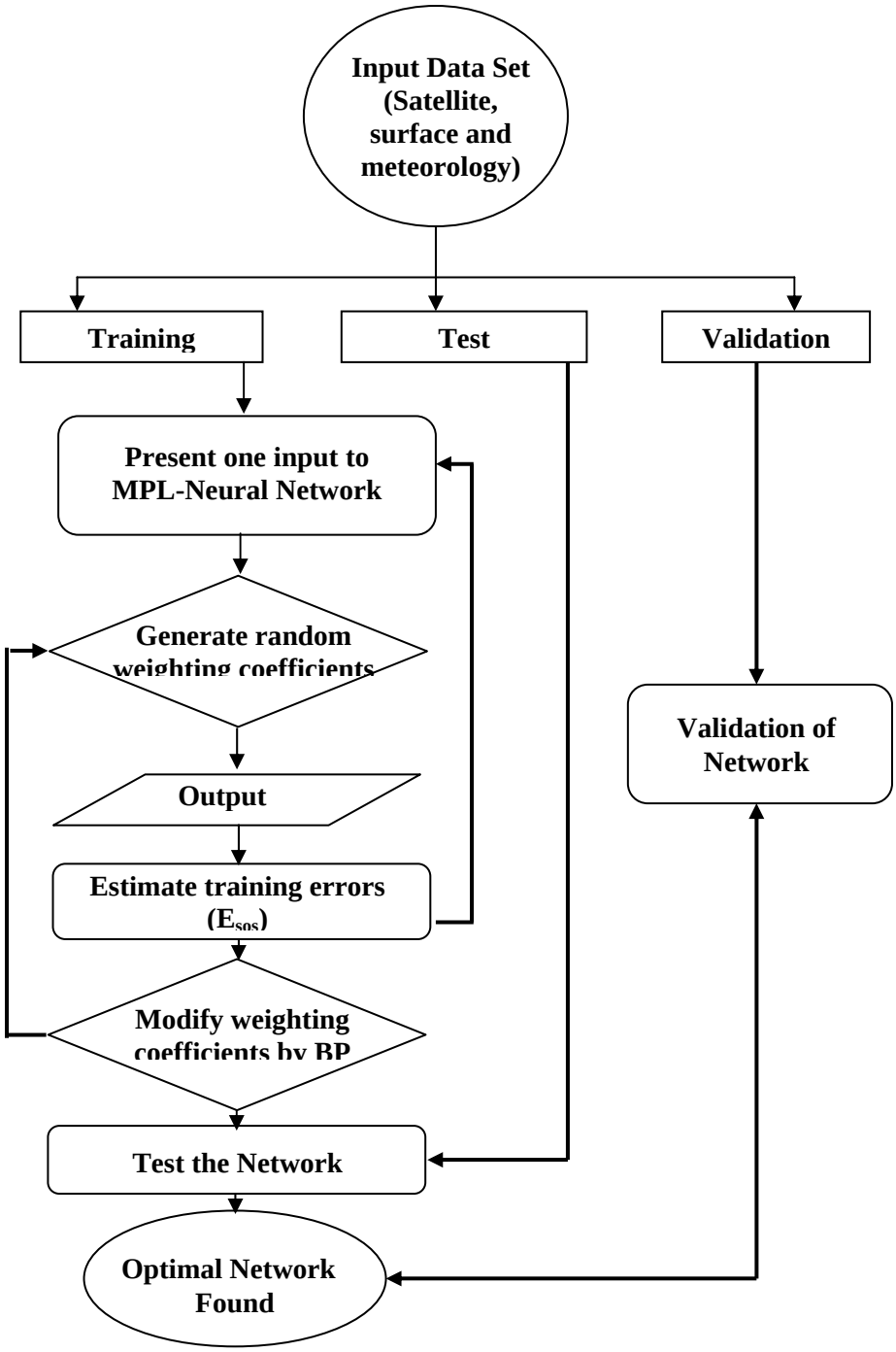

Figure 4.2 Flow chart describing training process of a multi layer perceptron neural network used for surface level particulate matter air quality assessment.

MODIS-Terra on 10x10 km^2 grid resolution have been used. All three data sets are first collocated in space and time using method described in *Gupta et al.,* [2008a]. Validation exercises [*Remer et al.,* 2005*; Levy et al.,* 2007] have shown that MODIS retrieve AOT over land within 10-20 % uncertainties. PM2.5 mass concentration measurements are made using a Tapered-Element Oscillating Microbalance (TEOM) instrument with an accuracy of ±1.5 μgm^{-3} for hourly averages. PM2.5 data were collected from 85 ground stations in Southeastern United States. Hourly analysis data of air temperature at 2 meter height (TMP), surface relative humidity (RH), wind speed at 10 meter (WS), and height of planetary boundary layer (HPBL) at 20x20 km^2 spatial resolution from RUC model are used in this study.

This integrated data base, which includes surface, satellite and meteorological information, contains 32834 samples. These large numbers of 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 neural network models the data are divided randomly into three subsets for training (50%), testing (10%), and validation (40%). Training data are used to train the MLP neural networks with multiple hidden layers. Twenty different combinations of neural network were trained for each model 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 number of hidden layers used. Number of hidden layers used in the model varies between 2 to 10 where error optimization is performed using sum of square of residual error function. Usually the use of more hidden layer makes

network size large, more difficult to train, thereby making it slower to operate while increasing the chance of over training. However, at the same time these networks perform better compared to networks with small number of hidden layers [*Bishop,* 1995].

Since, a large number of training samples (~15000) are available; we have allowed the number of hidden layers to vary from 2 to 10. Combination of linear activation functions (see section 4.2) are used in each of these 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. Multi layer perceptron uses a powerful second order back-propagation training algorithm, which converges quickly to solutions but requires large computer memory. Training of neural network is an iterative process. Weights coefficients associated with each node are modified using new data set after every iteration. After every cycle of training, test data sets were used to check the network's output with the desired output 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 entire training data sets. Equation 4.1 provides the formulation of SOS:

$$E_{sos} = \sum_{i=1}^{N} (Y_{tar_i} - Y_{out_i})^2 \cdots\cdots\cdots\cdots\cdots\cdots\cdots (4.1)$$

Where N is the number of training samples and Y_{tar} is the target value and Y_{out} actual output values of the ith sample. Figure 4.2 presents the flow chart describing entire training process.

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

$$APE = \frac{100 \times \left\langle \left| Y_{est} - Y_{obs} \right| \right\rangle}{\left\langle Y_{obs} \right\rangle} \cdots\cdots\cdots\cdots\cdots\cdots\cdots (4.2)$$

Y_{est} is estimated PM2.5 mass concentration (hourly or 24 hour 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 4.2) is also used to compare the results from neural network model with those obtained using two-variant and multi-variant regression models [*Gupta et al.*, 2008c].

4.4 Results and discussion

The goal of this paper is to assess the PM2.5 hourly and daily average mass concentration using MODIS AOT and meteorological fields from RUC. The model developed for this is based on a neural network and we have used MLP with different number of hidden layers and different activation functions. The results presented here are from the ensemble of five different models that performed the best out of the 20 models trained using training data sets. The current neural network has eight nodes (satellite and meteorological fields) in input layer with varying number of nodes in hidden layer and two nodes (hourly and daily average PM2.5 mass concentration) in 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 4.3. 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 inter-comparisons with TVM and MVM model outputs. PM2.5 from both training and validation data sets have been plotted as scatter plots. Separate analysis is performed for hourly and daily average PM2.5 mass concentration. We also compare the APE values obtained from neural networks with those obtained from two-variate models and multi variate models as described in *Gupta et al.,* [2008c]. The time series of observed and estimated PM2.5 mass concentration from the three approaches is also provided over selected stations.

4.4.1 Time Series Examples of Model Outputs

We evaluate the ANN over each station (total 85) by using predicted or estimated hourly and 24 hour average PM2.5 mass concentrations. Results obtained from ANN are also compared with those obtained using TVM and MVM as discussed in *Gupta et al.,* [2008c]. Figure 4.3(a-c) presents the time series of observed and estimated PM2.5 mass concentration (hourly) over three different stations. The three panels in figure 4.3(a-c) show the time series of hourly PM2.5 mass concentration estimated with TVM (top panel, red), MVM (middle panel, orange) and ANN (bottom panel, blue) along with

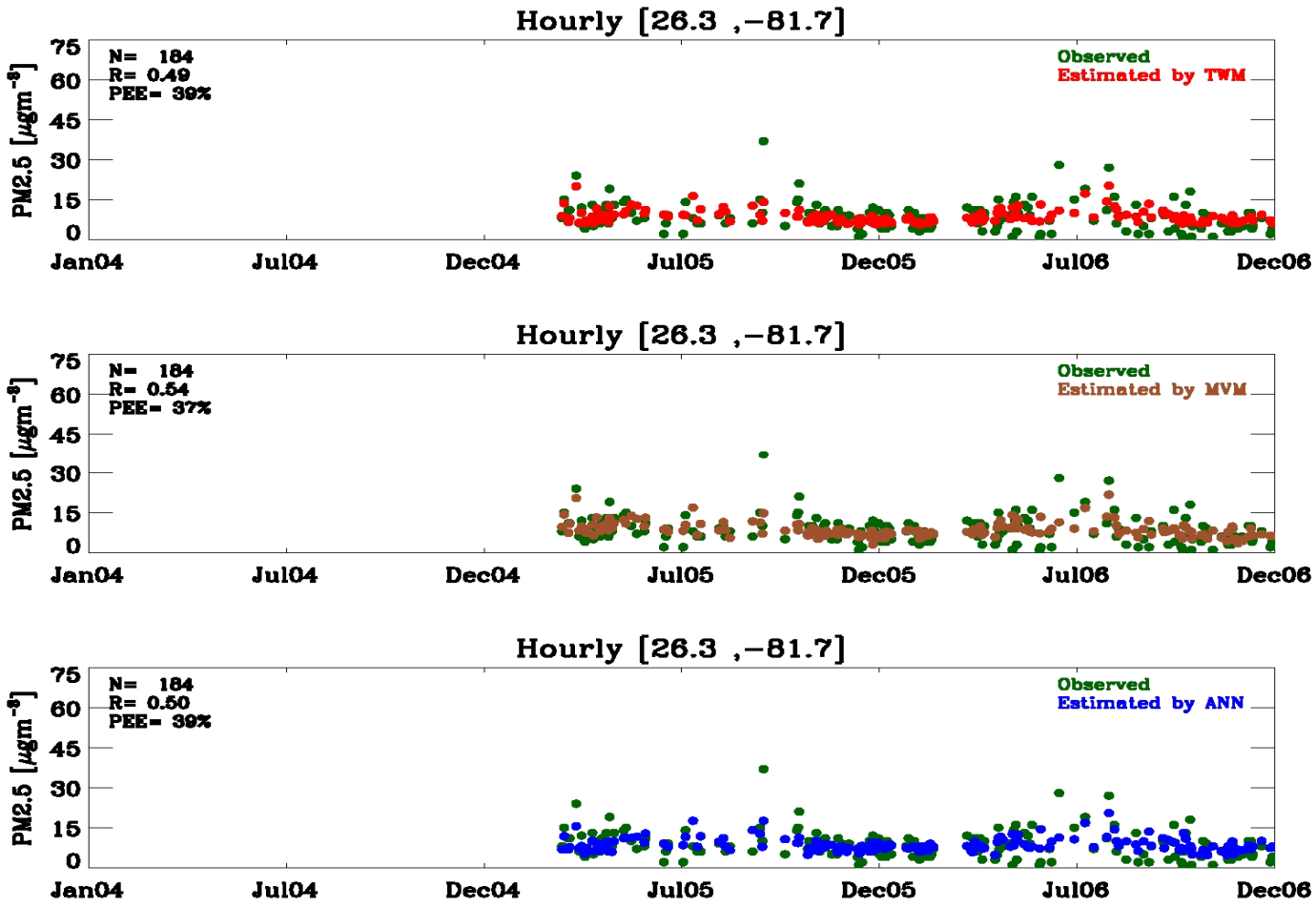

Figure 4.3a Time series inter-comparisons of PM2.5 mass concentration observed from surface station in and estimated using three different statistical models. Comparisons are between observed and estimate from two-variate (top panel), from multi-variate (middle panel) and from artificial neural network (bottom panel). This station is in Florida.

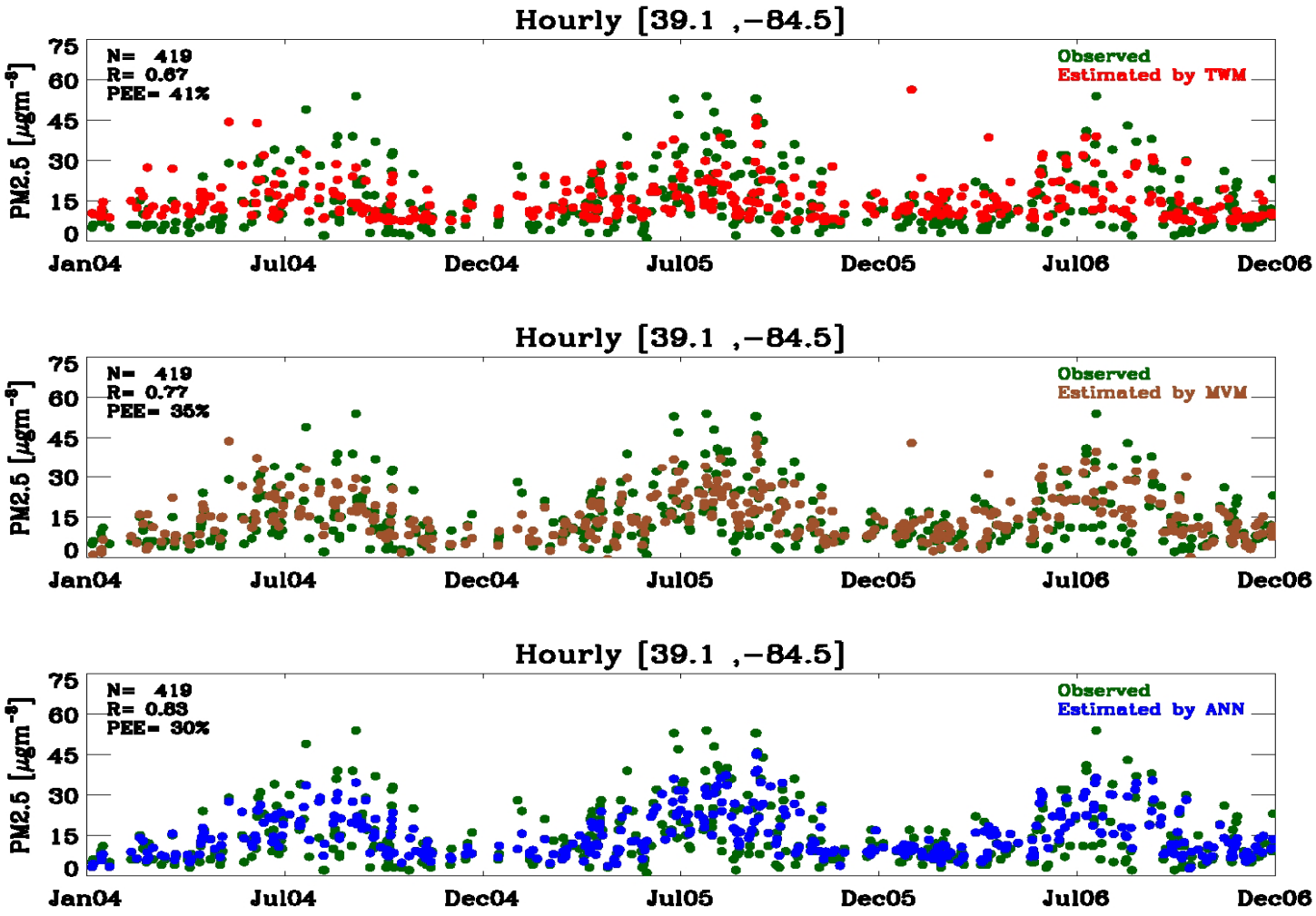

Figure 4.3b Time series inter-comparisons of PM2.5 mass concentration observed from surface station in and estimated using three different statistical models. Comparisons are between observed and estimate from two-variate (top panel), from multi-variate (middle panel) and from artificial neural network (bottom panel). This station is in Kentucky-Ohio border area.

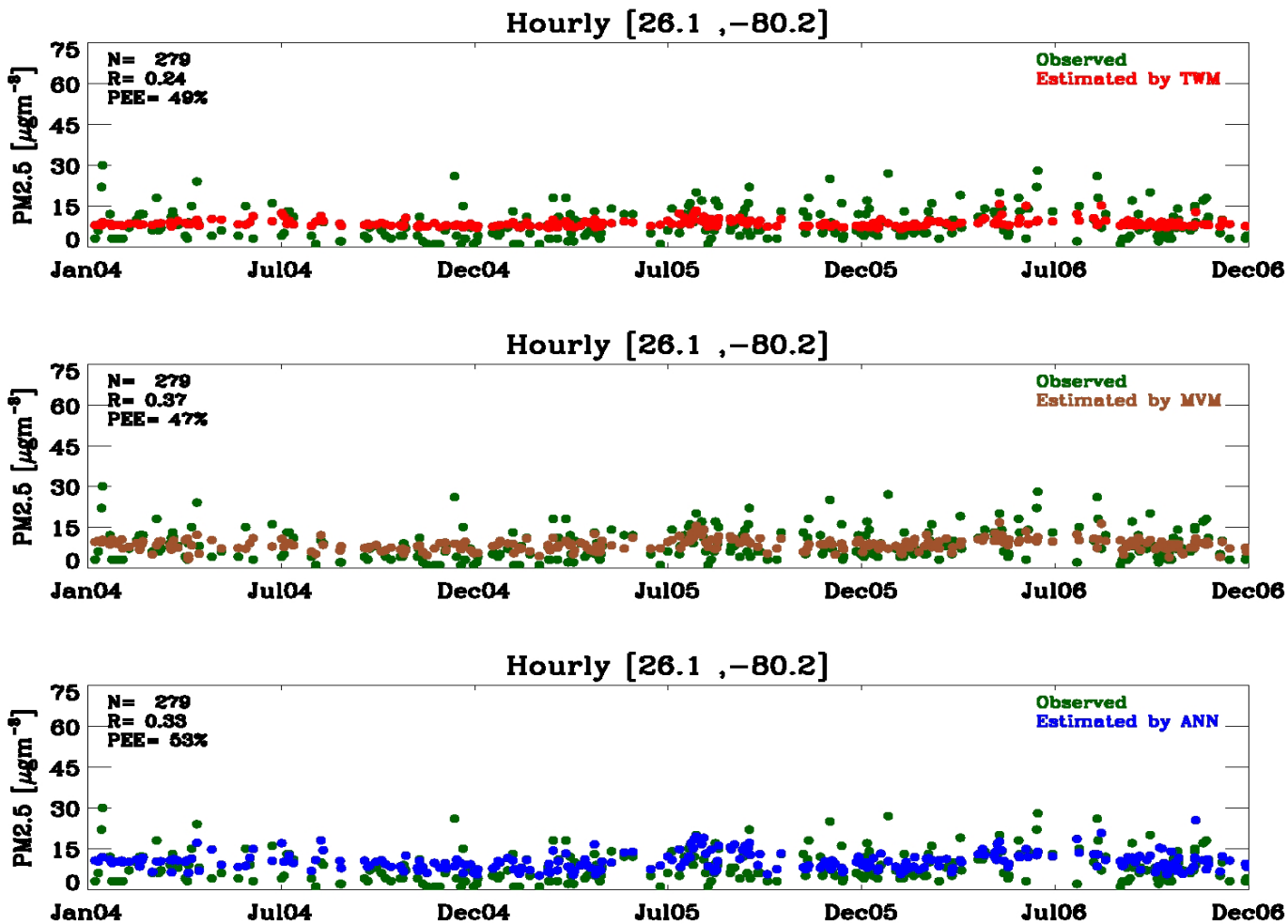

Figure 4.3c Time series inter-comparisons of PM2.5 mass concentration observed from surface station in and estimated using three different statistical models. Comparisons are between observed and estimate from two-variate (top panel), from multi-variate (middle panel) and from artificial neural network (bottom panel). This station is in Florida.

observed PM2.5 values (green). These stations have been chosen to demonstrate how well (or not) the ANN performs and how it compares with TVM and MVM.

Station number 1 (Figure 4.3a) is located in coastal area of Florida (26.3 N and 81.7 W; identify as station number 11 in *Gupta et al.,* 2008c). Use of ANN shows no improvement over other two methods while estimating PM2.5 mass concentration over this site. The correlations coefficient increased slightly from 0.49 for TVM to 0.50 for ANN whereas APE remained the same (39%). Multi-variate model, 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 performed very well for this station where variability in PM2.5 levels is also observed to be very low. Note that the main input component in these models is MODIS AOT, whose retrieval in coastal areas has larger uncertainties [*Remer et al.,* 2005; *Levy et al.,* 2008]. The time series of PM2.5 mass concentrations derived using all three methods does not follow the observed pattern and underestimate some high PM2.5 values as well as over estimating some very low PM2.5 values over this station. Similar results were observed for other coastal stations as well.

Figure 4.3b presents a similar time series analysis for a ground station on the Kentucky-Ohio border area (39.1 N and 84.5 W, station number 41 in *Gupta et al,* 2008c). This station is representative of other stations (>65%) where good degree of improvement in correlation coefficient and APE value is obtained while using ANN methods as compared to TVM and MVM models. Correlation coefficient increased from 0.67 for TVM to 0.77 for MVM and 0.83 for ANN. The APE values reduced to 30% for

ANN from 41% and 35% for TVM and MVM respectively. Figure 4.3b 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 6.2μgm^{-3}, 8.4μgm^{-3}, and 7.2μgm^{-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 concentration during pollution events with high PM2.5 values (PM2.5 >45 μgm^{-3}). This indicates that inclusion of local meteorology and satellite data sets in the model does improve the estimation of PM2.5 mass concentration but the degree of improvement varies with geographic locations.

Figure 4.3c shows an example of PM2.5 time series for a station where estimation accuracies degraded when ANN was used compared to MVM method. This is also a coastal station in Florida and is identified as station number 10 in *Gupta et al.,* [2008c]. Low level (<15μgm^{-3}) of PM2.5 mass concentration were observed during the study period with some exception when particulate matter air quality went to the moderate (>15.4 μgm^{-3}) category. The top panel in figure 4.3c clearly shows that the time series of estimated PM2.5 mass concentration using TVM is almost constant with very little 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%. But, use of ANN approach increased the error to 53%, although there is slight increment in correlation (0.33). Time series of these three stations exemplify the performance of ANN

approach and also aid comparison with the results obtained using TVM and MVM methods.

4.4.2 Evaluation of ANN and Comparison with TVM and MVM

Figure 4.4 presents scatter plots between observed and estimated hourly and 24 hour average PM2.5 mass concentration for both training and validation data sets. Also shown are best fit line, equation of this line, 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 hour averages except for a minor (≤0.01) change in slope values. Therefore, the ANN is producing almost similar results when using validation data sets which are not seen by the network during the training process. This identical performance of network on validation data sets shows the proper selection and distribution of training data sets, proper training process and generalizes the nature of the neural network trained. Our earlier study [*i.e., Gupta and Christopher,* 2008a] has shown that hourly PM2.5 correlated better with AOT as compared to 24 hour mean values of PM2.5 mass concentrations. But both MVM approach [*Gupta et al.,* 2008c] and ANN approach show that 24 hour average values are estimated more accurately by these models than hourly values of PM2.5 mass concentration. The reason behind this change is unknown but probably arises due to inclusion of meteorology in the analysis. 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 (~6.2) shows the mean bias in the estimated mass concentrations. In all four panels the higher values of PM2.5 are

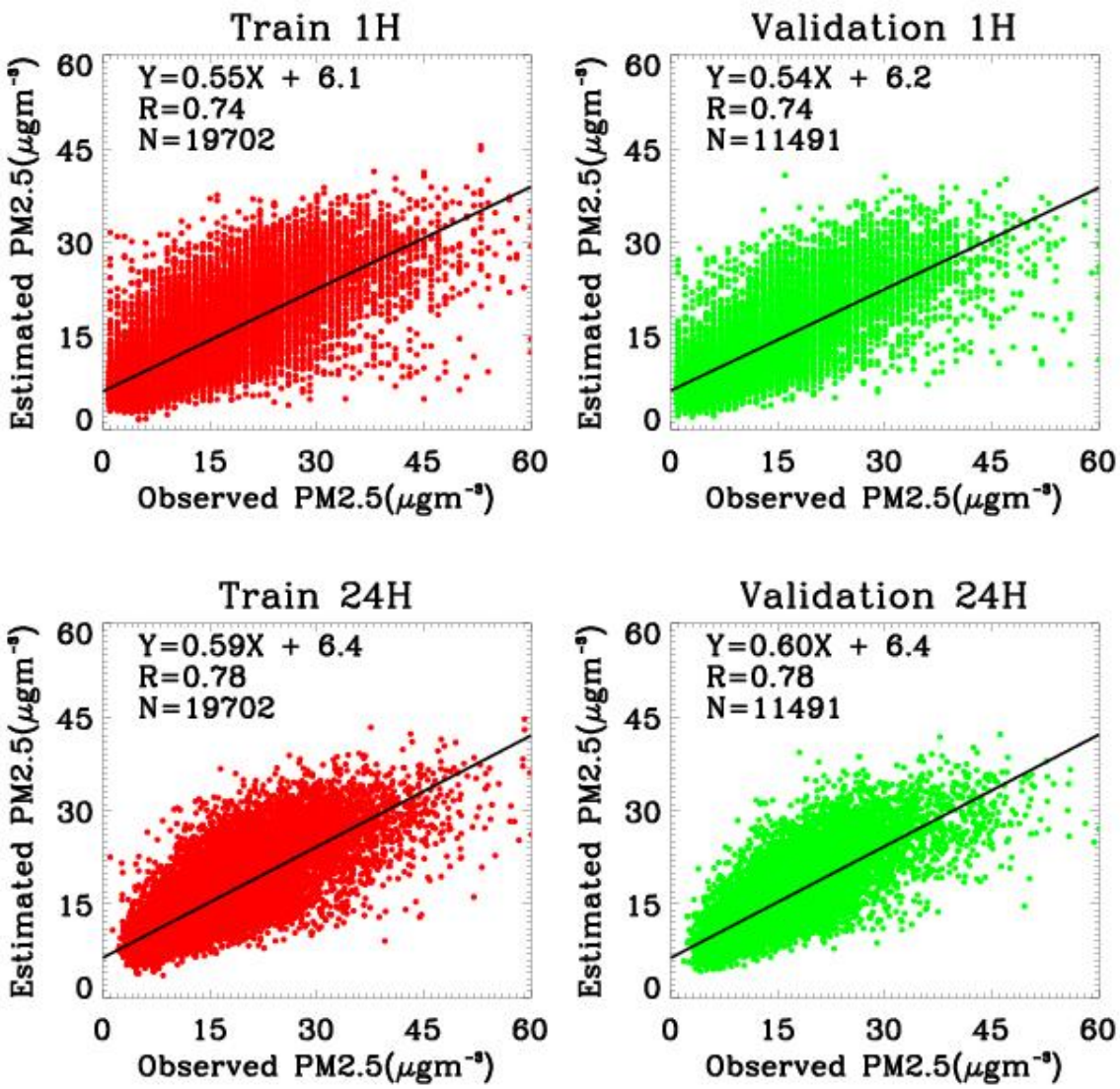

Figure 4.4 Scatter plot showing performance of trained neural network on all the data sets over three year time period. Separate plots are presented for hourly (top panels) and daily average (bottom panels) PM2.5 values as well as for training (left, in red color) and validation (right in green color) data sets.

underestimated and in some cases overestimation occurs in lower range of PM2.5. Multi-variate method also produced underestimation of PM2.5 mass concentration in the higher range. Earlier speculation for this type of model behavior was small number (<1% of total samples) of available samples for high (>45μgm^{-3}) PM2.5 mass concentrations [*Gupta et al.,* 2008c]. For further analysis, relation between only high PM2.5 and corresponding AOTs have been shown in Figure 4.5. Figure 4.5 clearly shows that there are many high PM2.5 values between 45 μgm^{-3} and 60 μgm^{-3} for which AOTs vary all the way from almost 0.0 to 1.4. Small range of PM2.5 mass concentration loading 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). This type of AOT-PM2.5 behavior indicates the possibility of multiple layers of aerosols (PM2.5) in the atmosphere. In such cases, aerosols that are aloft could also contribute towards the total columnar AOT values and hence columnar AOT does not show a good agreement with surface level loading of aerosols. Specific pollution episodes such as biomass burning can produce such multiple layers of aerosols [*Jones and Christopher et al.,* 2008] aloft up to 5 km in the atmosphere. Vertical aerosol layer information from lidars (such as from CALIPSO) are required to understand this issue. Results from this network using all the data sets provide almost 15% and 21 % reduction in error (APE) compared to TVM model in *Gupta et al.,* [2008c], 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.

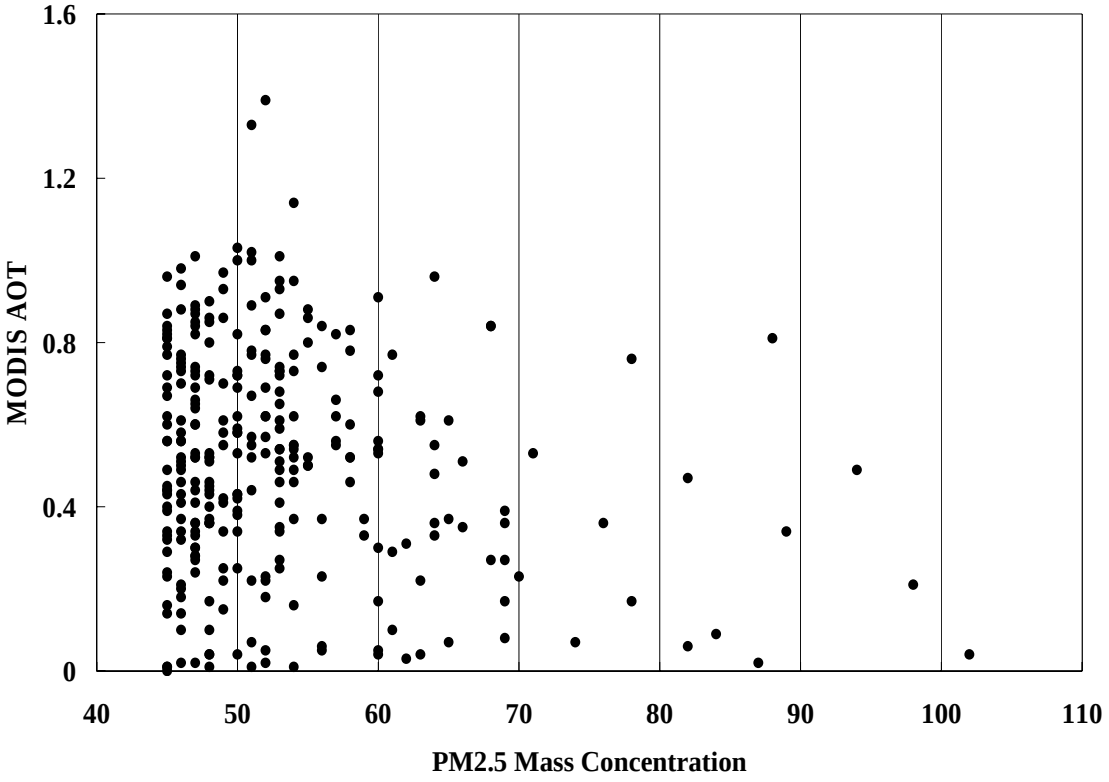

Figure 4.5 Scatter plot between MODIS AOT and PM2.5 mass concentration (hourly PM2.5>45μgm^{-3}) showing the poor AOT-PM2.5 relationship under high pollution events.

In order to understand the seasonal behavior of neural networks, entire data sets are separated into four seasons. Data for each season is again separated into training, validation and testing data sets (section 4.3). Training data set for each season are used to train four different networks exclusively for each season. Figure 4.6 (a-h) presents the results from validation exercise of seasonal networks developed exclusively for each season. Correlation coefficients for hourly average PM2.5 estimations are highest (0.76) in fall and lowest (0.49) in winter seasons whereas it is 0.70 and 0.63 in spring and summer. Poor estimation during winter months are associated with very low HPBL and AOT values, which are subject larger uncertainties. Whereas in summer months planetary boundary layer is well mixed and is deeper, and we have shown in previous study [*i.e., Gupta et al.,* 2008c] that AOTs are well correlated with surface level pollution under high HPBL. R values in case of 24 hour average for winter, spring, summer and fall are 0.57, 0.73, 0.67, and 0.82 respectively. Network behavior in different season is almost same as MVM but estimation accuracies are improved. Absolute percentage error of estimation has improved being highest (23%) in fall and lowest (12%) in summer and intermediate values in winter (16%) and spring (18%) season. The improvement in APE value is higher in case of 24 hour average PM2.5 mass concentration estimations (table 4.2).

Table 4.2 and 4.3 provides inter-comparison of three different approach of estimating PM2.5 mass concentration using satellite data sets. The statistical parameters represent the results from evaluation of these methods are absolute percentage error of estimation (APE) and linear correlation coefficients (R). Numbers in bracket in table 4.2 and 4.3 represent the improvement in the statistical parameter over two variate method (TVM). The highest improvement in R value for hourly PM2.5 is in winter (133%)

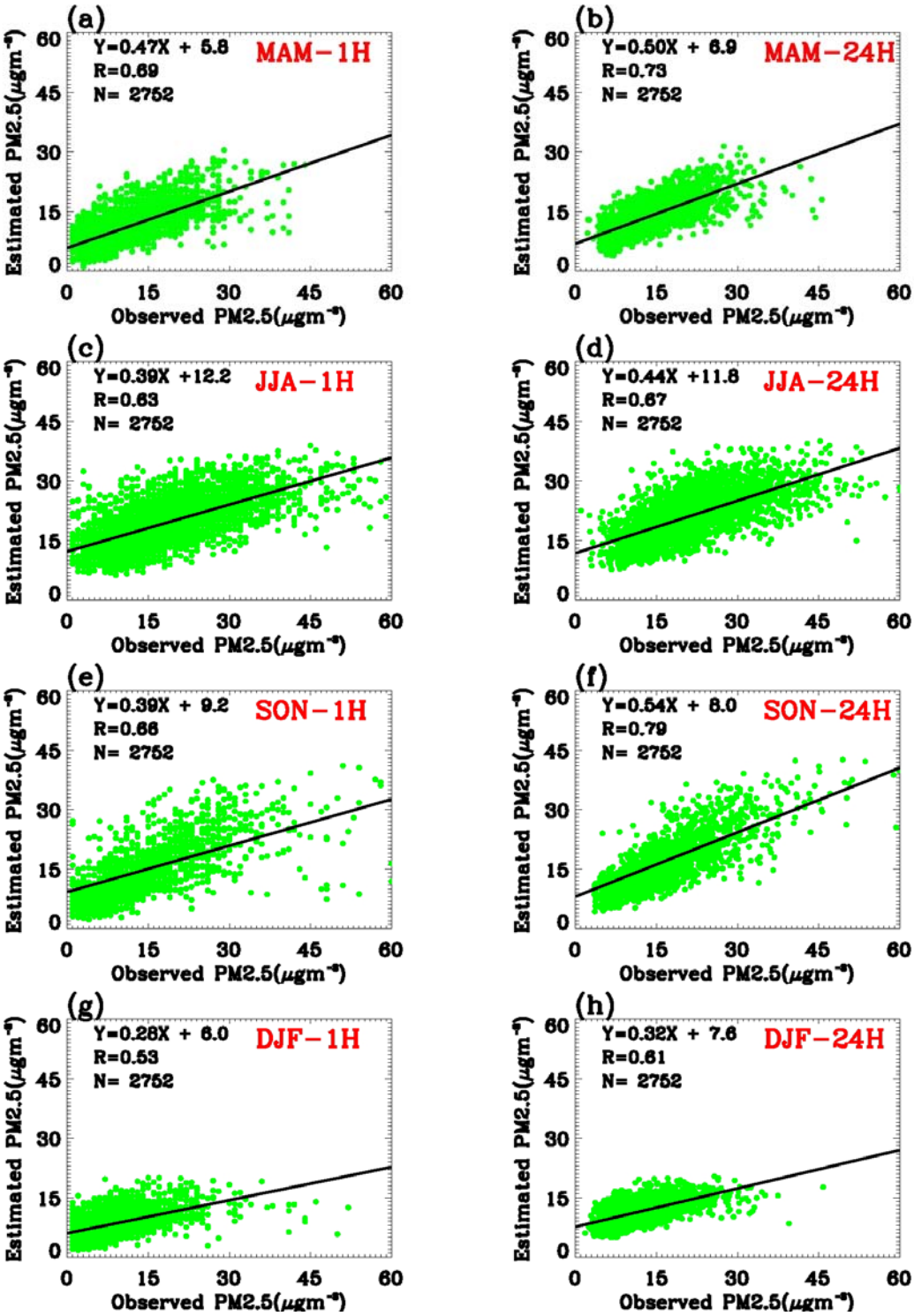

Figure 4.6 Validation of neural network models trained for each season using training data sets. Scatter plot between observed and estimated PM2.5 mass concentration for hourly and daily average are presented here.

Table 4.2 Absolute percentage error (APE) matrix for three different statistical approaches used to estimate surface level PM2.5 mass concentration using satellite remote sensing data sets.

Serial Number & Data		**Absolute Percentage Error (APE)**					
		Model Type					
		Two Variant		**Multi-Variant**		**Neural Network**	
		1 Hour	***24 Hour***	***1 Hour***	***24 Hour***	***1 Hour***	***24 Hour***
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)

Table 4.3 Linear correlation coefficient (R) for three different statistical approaches used to estimate surface level PM2.5 mass concentration using satellite remote sensing data sets.

Serial Number & Data		**Linear Correlation Coefficient (R)**					
		Model Type					
		Two Variant		**Multi-Variant**		**Neural Network**	
		1 Hour	***24 Hour***	***1 Hour***	***24 Hour***	***1 Hour***	***24 Hour***
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)

whereas lowest improvement is in fall (23%) season. Similarly, improvement in R for 24 hour average PM2.5 is highest in winter (256%) and lowest in fall (32%). It is clear that 24 hour average PM2.5mass concentration estimation shows more reduction in error compared to 1 hour average PM2.5 in all conditions.

Figure 4.7 shows the validation of ANN system at a specific station (station number 72). This is the same stations used in part I of this study to demonstrate the MVM model performance. This station represents the cases where PM2.5 estimation using new model improved significantly. Further details on these stations can be found in *Gupta et al.,* [2008c]. The improvement over the station is significant and apparent. Correlation coefficient has changed by 18% i.e. from a value of 0.73 to 0.86. The percentage error of estimation for this station is 29% and 18% for hourly and 24 hour average PM2.5 mass concentration.

4.5 Summary and Conclusions

Information on surface level PM2.5 mass concentration is very useful for monitoring and regulating particulate matter air quality. Satellite data is 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. The derivation of surface level PM2.5 mass concentration 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 concentrations at surface level. In the current study we explored the possibility of using an artificial neural network system for estimating PM2.5 mass concentration instead of simple regression equations.

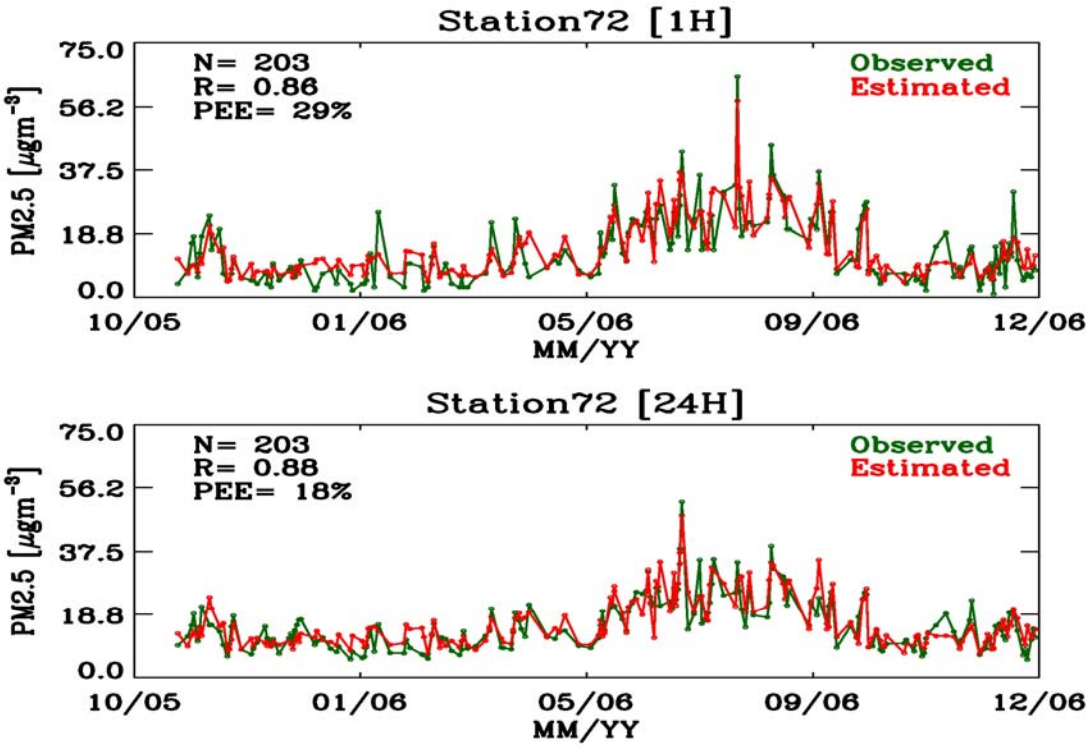

Figure 4.7 Example of time series validation of model based on neural network for hourly and daily average PM2.5 mass concentration estimation over station in Tennessee.

Several neural network models have been trained, tested and validated using three years of surface, satellite, and meteorological fields in South Eastern United States. This artificial neural network based model takes satellite derived AOTs and model produced meteorological fields as input and estimates hourly and daily averaged PM2.5 mass concentrations. We also compare 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 correlations 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 concentrations respectively. Further analysis shows that improvements in R and APE values vary with seasons as well as geographical locations. Fall months show highest improvement in APE and R values for ANN compared to TVM and MVM models. Absolute percentage errors for fall months are improved highest 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 shows 6% to 8% improvements in APE values over MVM models. Improvement in correlations during winter season is almost two to three folds for ANN over TVM. These improvements in R and APE values are even higher over some stations whereas similar over most of the stations. Overall ANN shows more improvement in accuracies for daily averaged PM2.5 mass concentration estimation than hourly averaged values, which is similar to MVM method. 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 concentration during high pollution level at surface as well as aloft in the atmosphere presents more challenges for all three models. High aerosol loading 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 lidar (CALIPSO) should improve our understanding and accuracies of PM2.5 estimations. Artificial neural network underestimated high PM2.5 mass concentration, which makes them similar to MVM models in current study. Special treatment and more research are 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.

CHAPTER 5

SUMMARY AND CONCLUSIONS

5.1 Discussion

To understand the effects of particulate matter on the earth's climate system and on human health, it is necessary to routinely monitor $PM_{2.5}$ on a global basis. This task is challenging because these sub-micron aerosols are highly variable in space and time. Typically, using ground-based instruments, PM2.5 mass concentration of ambient particles is widely measured in both urban and rural areas of Europe, United States, Australia, and in some parts of Asia for monitoring PM2.5. However, these ground-based observations represent point measurements and do not have the necessary spatial coverage to map the regional to global distribution of PM (aerosols). On the other hand, daily satellite observations that provide near global coverage, can serve as a surrogate for monitoring PM2.5 air quality.

Recent studies using satellite data from MODIS [*Chu et al.,* 2003*; Wang and Christopher.,* 2003*; Gupta et al.,* 2008a] and MISR [*van Donkellar et al.,* 2007*; Liu et al.,* 2008a] have shown the tremendous potential for mapping global distribution of aerosols and their properties and for deriving surface estimates of particulate matter, particularly PM2.5.

The potential for monitoring PM2.5 air quality using satellite data from space based sensors for regional to global scales have been recently demonstrated using aerosol optical thickness data products [*e.g. Wang and Christopher*, 2003*; Chu et al.*, 2003*; Engel-Cox, et al.*, 2004*; Al-Saadi et al.*, 2005*; Liu et al.*, 2008a*; Gupta et al.*, 2007*;* 2008a; 2008b]. The AOT is the integral of atmospheric extinction from the surface to the top of the atmosphere. This columnar retrieved parameter is dependent on aerosol mass concentration, mass extinction efficiency, hygroscopic growth factor, and effective aerosol scale height. Thus, AOT that includes information of aerosol mass concentration can be related to $PM_{2.5}$ and can be used to infer air quality [*Wang and Christopher*, 2003].

Although the satellite based retrieval of air quality is promising, it poses several challenges. There are many factors that can affect the relationship between AOT and $PM_{2.5}$. For example, the satellite-derived quantities provide columnar information for ambient conditions whereas the $PM_{2.5}$ measurements are representative of near-surface dry mass concentrations. The satellite footprints represent large spatial areas and are subject to cloud contamination [*Zhang et al.*, 2005]. Other issues including variations in aerosol type and hygroscopicity must also be adequately understood before using satellite data for air quality assessment.

In our previous studies [i.e., *Gupta et al.*, 2006; 2007], using regression analysis, we derived empirical relationship between 24 hour $PM_{2.5}$ mass concentration (μgm^{-3}) and MODIS AOT (at 0.55 µm) and concluded that the satellite derived AOT is an excellent tool for air quality studies over large spatial area. Our study confirmed previous regional and global studies over limited locations [*e.g. Wang and Christopher*, 2003*; Chu et al.*,

2003]. We have performed this analysis over various locations in United States, India, Australia, Hong Kong and Switzerland.

This dissertation goes beyond establishing the AOD-PM2.5 relationships to estimate particulate matter air quality and addresses in detail other factors such as the influence of meteorological parameters that governs this relationship. One of the major factors that limit the usefulness of the AOT-PM2.5 relationship is the lack of information on the vertical distribution of aerosols. Since AOT is a columnar quality it is important to know what fraction of aerosols is being 'sensed' at the ground. Obviously, using lidar data sets from space or ground will be of tremendous help. However, currently these data sets are not available on a daily basis. Therefore the use of mixing heights derived from meteorological parameters to address mixing layer heights is one of the focus areas of this dissertation.

Since, satellite data are used to assess PM2.5 air quality, the quality of the data sets must be assessed. In Chapter 2, a rigorous assessment of the satellite data used in this study was presented [*Gupta and Christopher,* 2008a]. Since air pollution from satellites can only be assessed in cloud free conditions, a sampling bias is induced in the inferred PM2.5 concentrations. Appendix A provides a complete assessment study of this sampling bias [*Gupta and Christopher,* 2008b]. Comparisons between the prior two variate methods and a new multi-variate method based on satellite, ground-based and meteorological information was presented and discussed in Chapter 3 [*Gupta et al.,* 2008c]. Finally an ANN based system for assessing PM2.5 is discussed in Chapter 4 [*Gupta et al.,* 2008d]. The key results from this dissertation are as follows:

1) Seven years of the MODIS-Terra AOT data and PM2.5 mass concentration from ground measurements over one site (North Birmingham, 33.55 N, 86.82 W) in the Southeastern United States have been used to perform a comprehensive analysis of various aspects of particulate matter air quality [*i.e, Chapter 2; Gupta and Christopher,* 2008a]. Our results indicate that PM2.5 mass concentration over Northern Birmingham has decreased by about 23% in year 2006 when compared to year 2002 and that the air quality during summer months is poor when compared to winter months. The correlation between PM2.5 and MODIS AOT increased from 0.52 to 0.62 when hourly PM2.5 data were used instead of daily mean PM2.5 data. Changing the grid size used for averaging the satellite data around the ground station during comparisons produced less than ±0.03 difference in mean AOT values for 90% of the observations. Application of AOT data quality flags reduces the sample size but does not affect AOT-PM2.5 relationship significantly. We recommend using AOT data quality flags for daily analysis, whereas long time scale analysis can be performed using all the AOT retrievals to obtain better sampling. This conclusion is especially useful to modeling communities that are assimilating satellite data products for forecasting PM2.5.

2) Although satellites provide reliable and repeated measurements on a global scale, particulate matter air quality information can be derived from satellites only when clouds are absent and when surface conditions are favorable. However, ground measurements provide particulate matter information irrespective of cloud cover and surface conditions. Therefore there could be a sampling bias when using clear sky satellite data for air quality research. To examine this issue, we calculate average particulate matter (PM2.5) mass concentration from daily ground-based measurements

(ALLPM) on monthly to yearly time scales and compare these against the same ground-measurements but averaged for only those days when satellite data is available (SATPM). Results indicate that satellite data are generally available less than 50% of the time over these locations, although the inter-regional variability of data availability is from 32% to 57 %. However, the mean differences between the ALLPM and SATPM, over monthly to yearly time scales over the Southeastern United States, is less than 2 μgm^{-3} indicating that low sampling from satellites due to cloud cover and other reasons is not a major problem for studies that require long term PM2.5 data sets. These results have important implications for satellite studies especially over areas where ground-based measurements are not available. This conclusion and analysis methods are useful for epidemiological communities that are beginning to explore the potential of using satellite data for long term exposure studies.

3) The vertical distribution of aerosols and meteorological parameters such as wind speed, temperature and humidity also play a major role in this AOT-PM2.5 relationship. Our results indicate that there are up to three fold improvements in the correlation coefficients when using multi-variate regression as compared to the two-variate regression of AOT and PM2.5 concentration. A 20-50% improvement in RMSE is observed when temperature and boundary layer height is added to the AOT-PM2.5 relationship. Since boundary layer heights are readily available from model simulations, they can be used as a good surrogate for estimating aerosol heights in conjunction with space and ground-based lidars. This conclusion is useful for communities that are utilizing AOD-PM2.5 relationships on a near real time basis for assessing PM2.5 from satellites.

4) Use of an artificial neural network framework for assessing particulate matter air quality has also been explored in this study. Several neural networks have been trained as a function of season and different restrictions on the data sets to estimate PM2.5 mass concentration for hourly and daily particulate matter air quality monitoring. These ANN based models are validated using separate validation data sets not used in the training of the NN system. The results from ANN show an improvement in the correlation coefficients from 0.78 to 0.85 when compared to two-variate and multi-variate regression equations. The improvement in the absolute percentage error of estimation ranges from 5% to 45% over different seasons and regions when compared with the two-variate models. While the use of the ANN has been exploratory in this dissertation, it appears that this method holds promise for future work to estimate PM2.5 based on forecasted meteorological fields.

Finally, this research has shown that MODIS AOTs can serve as a surrogate to assess surface level PM2.5 mass concentration and the use of meteorology further improves this relationship. This comprehensive study using multi-year data sets over 85 different stations in south east United States clearly shows that there is 5-50% improvement in estimation accuracies while using MVM and ANN approach over TVM approach.

5.2 Future Work

As a follow on to this dissertation, these three methods of estimating PM2.5 mass concentration using satellite data will be tested in other areas around the United States and the world. Global mega cities air quality will also be evaluated using multi

year satellite data sets and the methodologies developed during this dissertation. Issues related to underestimation of high PM2.5 values will be analyzed in more details and adjustment to the models will be performed to improve estimation of high PM2.5 values.

APPENDIX A

EVALUATION OF SAMPLING BIAS IN SATELLITE DATA

A.1 Introduction

Particulate matter (PM) is a mixture of both solid and liquid particles suspended in air and is usually classified as fine (PM2.5, d<2.5 µm) and coarse (PM10, 2.5<d<10 µm), where d is the aerodynamic diameter. In this paper, we are primarily concerned with PM2.5 that could be from various sources including dust, vehicle and industrial emissions, forest and agricultural fires. PM2.5 air quality continues to degrade throughout the world due to increasing pressures of urbanization that has serious implications for health, climate, visibility, and hydrology [*Kaufman et al.,* 2002]. Although some countries have a dense network of PM2.5 monitoring stations [*Al Saadi et al.,* 2005], worldwide, there are limited ground-measurements thereby creating a challenge for monitoring and studying air pollution. With the launch of Terra and Aqua polar orbiting satellites, there has been an increased emphasis for using satellite data to study PM2.5 to alleviate some of the problems due to the unavailability of ground measurements [*Al Saadi et al.,* 2005]. While satellites can provide reliable, repeated measurements from space, monitoring surface level air pollution continues to be a challenge since most satellite measurements are column-integrated quantities. However, several studies have

shown that satellite data can be a good surrogate for ground measurements provided appropriate adjustments are made for converting columnar quantities to surface values [*van Donkelaar et al.,* 2006*; Liu et al.,* 2004]. With new satellites that can now provide vertical distribution of aerosols and clouds, we are poised to make significant advances in using satellite data for particulate matter air quality research [*Engel-Cox et al.,* 2006].

The link between PM exposure and adverse health recently prompted the United States Environmental Protection Agency (EPA) to tighten its 24-hour fine particle standard from 65μgm^{-3} to 35μgm^{-3} [*Fed. Reg.,* 2006]. Studies show that long-term particulate matter exposures are associated with death due to heart failure, and cardiac arrest [*Pope et al.,* 2002]. However it is difficult to obtain long term estimates in large spatial scales from the limited number of ground measurements and therefore the use of satellite data could be beneficial.

Several research papers have outlined the methods by which satellite data can be used to obtain surface PM2.5 [e.g., *Wang and Christopher,* 2003*; Engel-Cox et al.,* 2006*; Hutchison et al.,* 2005*; Gupta et al.,* 2006*; Liu et al.,* 2004*; van Donkelaar et al.,* 2006]. In summary, first the columnar satellite-derived aerosol optical Thickness (AOT) values are related to surface PM2.5 mass measurements. Then this AOT-PM2.5 relationship can be used to convert the satellite measurements to air quality indices based on EPA guidelines. These values are then color coded for dissemination to the public where Green is for Good air quality and Orange and Red are poor quality. A good example of this can be seen at http://alg.umbc.edu/usaq/.

Given the links between PM2.5 and health, and the scarcity of monitoring stations throughout the world, satellite remote sensing appears to be the only viable method to

monitor PM2.5 air pollution over large spatial scales. However, satellite retrievals of AOT rely on cloud-free conditions and favorable surface conditions to obtain PM2.5 air quality thereby limiting the number of days where satellite data can be used over a certain location. Also satellite retrievals are sometimes not available due to various retrieval issues such as bright surface backgrounds and data dropouts. What the satellite retrievals lose in terms of cloud cover limitations, it makes up in terms of the wide spatial coverage that is often useful for assessing how the pollution plumes move from one area to another [*Hoff et al.,* 2005]. Even with these limitations, satellite data sets due to their global coverage are a valuable asset for monitoring PM2.5 air quality [*Gupta et al.,* 2006].

In contrast, ground measurements of PM2.5 are available regardless of cloud cover and depending upon the location, measurements are made available every hour or as 24-hour averages. While this is extremely useful, ground measurements are limited due to lack of spatial coverage or unavailability. Since continuous monitoring of PM2.5 is essential and monthly and annual averages of PM2.5 air quality is vital for assessing global air quality, it is important to assess whether satellite data can provide the sampling necessary to monitor air quality over these time scales. We assume that satellite data sets are good surrogate for monitoring surface PM2.5 air quality while recognizing that there are indeed some research limitations that are currently being addressed using new satellite data, meteorology, and other tools [*e.g. Engel-Cox et al.,* 2006].

Since PM2.5 mass is measured from the ground irrespective of cloud cover while satellite data only provide AOT information during cloud-free and favorable retrieval conditions, we ask the following questions, ‘What is the difference between ground-based PM2.5 (ALLPM) and the PM2.5 for only those days where satellite data are

available (SATPM) on monthly and yearly time scales?' How many days of satellite data are available due to cloud cover contamination and other limitations for PM2.5 air quality research? Understanding these differences are important to address the utility of satellite data in mapping PM2.5 air quality over monthly and yearly time scales especially since long term exposure studies require global data sets on yearly time scales [*Pope and Dockery,* 2006]. Note that we are not using the satellite-derived AOT in this paper; rather we simply examine the PM2.5 during the time of the satellite overpass. To examine this issue, we selected the EPA region 4 in Southeast United States (Figure A.1) where previous research has shown that satellite data is indeed a robust surrogate for PM2.5 estimation [*Wang and Christopher,* 2003]. This region was also selected due to the numerous ground-based PM2.5 measurements that are available to address the aforementioned questions.

A.2 Data and Method

We obtained 24-hr PM2.5 mass concentration values from 38 ground monitoring stations in Southeastern United States from February 24, 2000 to December 31, 2005 covering eight states in EPA region 4. Figure A.1 shows the location of air quality stations used in the current study. We used these PM2.5 values to calculate monthly, seasonal and yearly averages (ALLPM). We then obtained six years of the MODIS satellite data [MODO4, V005] [*Levy et al.,* 2007] that contain AOT and other geophysical parameters in 10 km^2 grid resolution. MODIS AOT is retrieved for cloud-free conditions and when surface reflectance in the 2.1 μm channel is less than 0.4.

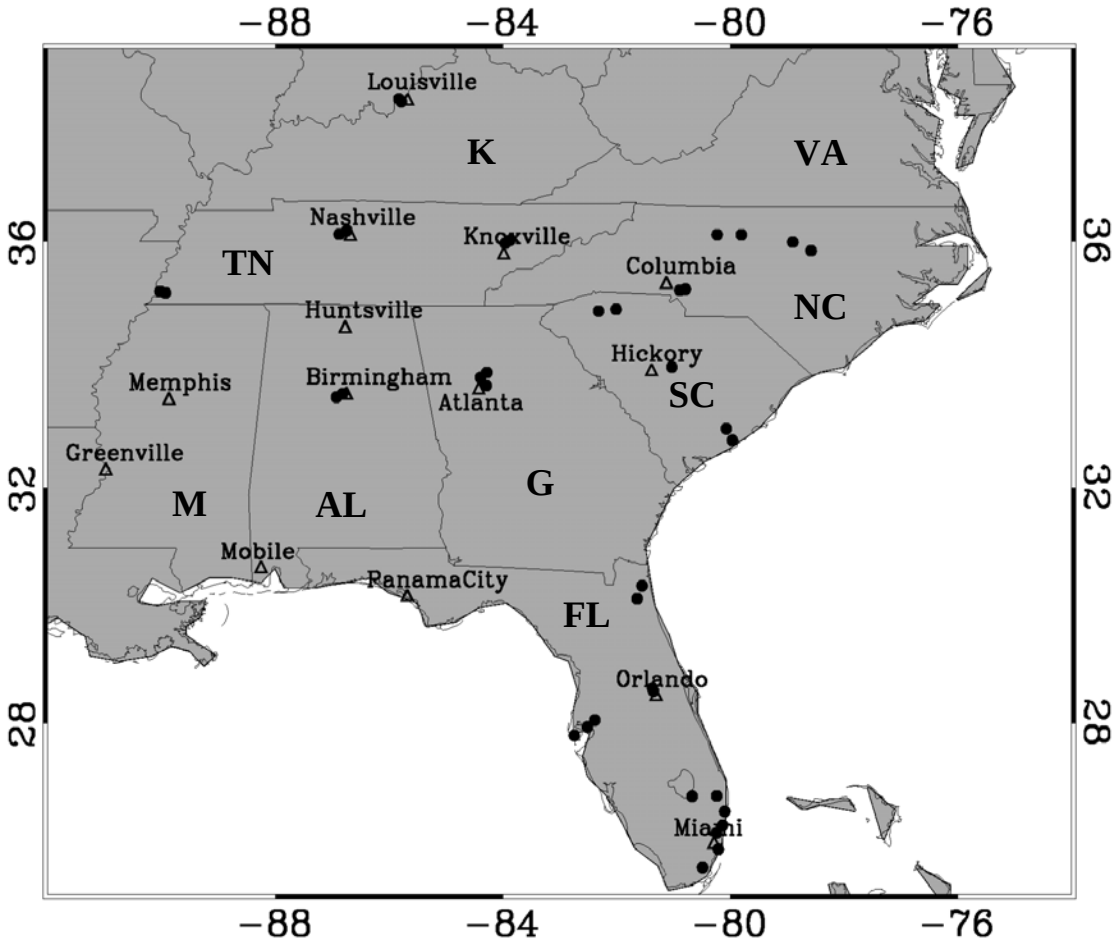

Figure A.1 Area of study and location of PM2.5 measuring stations in the South Eastern United States.

The MODIS algorithm also considers the retrieved AOT as questionable if 2.1 μm channel reflectance is more than 0.25 [*Levy et al.,* 2007]. For each one of the 38 PM2.5 stations, a 5X5 group of the 10 km^2 pixels centered on the ground station are examined. This method of using 5X5 pixels is often the standard practice when comparing ground based with satellite measurements [*e.g. Ichoku et al.,* 2002]. If MODIS retrieved AOT is present on any given day, over the ground location, and then the PM2.5 value from the ground was included in computing monthly, seasonal, and yearly values of PM2.5 mass. Note that the satellite-derived AOT values are not used in the calculations. We only use the satellite data to check if AOT retrievals are available for the 5X5 pixels grid. Even if one of the pixels in the 5X5 grid had a reported AOT value, the ground-based PM2.5 for these days are tagged and labeled as SATPM, since this is what the Terra-MODIS will sample over time. In this way, we measure the difference between mean PM2.5 values from the all ground measurements (ALLPM) and PM2.5 values from the only those ground measurements when the satellite derived AOT values are available (SATPM). We also track the number of days when satellite data are available in a given month, season, or year which is called SAT DAYS through out the paper. For example, if a 5X5 grid had reported AOT values for every single day in an entire month, then it would indicate that there was 100% data availability from the satellite during that month. However, during the six year period, only 32-57 % of data (SAT DAYS) is available over the various locations (Figure A.3). Many stations in Florida and North Carolina are located near the coast and therefore fewer SATDAYS are available due to MODIS AOT retrieval limitations in coastal regions [*Remer et al., 2005*]. For all of the following analysis we set 85% thresholds on data availability from the surface data, to maintain uniformity

across all locations. For annual analysis, we used data from all locations if they had greater than 310 out of 365 days of data, for seasonal analysis 75 out of 90, and for monthly analysis 25 days of 30 corresponding to about 85% for each case. Total 74876 data points from the surface and 33211 data points from the satellite are used. Table 1 provides geographical locations and other detailed information and it also includes ALL DAY, SATDAY, mean PM2.5 mass concentration and mean AOT value for each station. Daily mean values are averaged over the entire study period.

A.3 Results and Discussion

We first examine the ratio of ALLPM to SATPM for 71 months. In ideal conditions, ALLPM/SATPM for every ALLPM value should be a straight line centered at 1.0. Any deviation from this 1.0 value will represent a bias due to the low sampling by MODIS. A scatter plot of the ratio between ALLPM and SATPM as a function of ALLPM is shown in Figure A.2. The mean and standard deviation in the ratio of ALLPM to SATPM is 0.96±0.15 with very few points (28) having ratios greater than 1.5 indicating that 99% of the time the SATPM has similar PM2.5 values as ALLPM, with small overestimation (<1%) by SATPM. It is also important to note that high ALLPM/SATPM values correspond to ALLPM values less than 10μgm^{-3}. Further analysis reveals that all these high ratio values correspond to 13 different stations during 2000, which could be due to partial or full data loss during MODIS initial phase of data collection. The highest value (3.9) of this ratio occurred in May 2000 when satellite data only exist 7 out of 31 days for that particular station. To examine these differences in detail, both ALLPM and SATPM is calculated for each station separately.

Table A.1 Detailed information on surface locations of PM2.5 mass measurements along with mean values of PM2.5, MODIS AOT and available number of days.

Station No.	City, State	Lat.	Lon.	Months	ALL	PM2.5	SAT days	AOT
1	Birmingham 1, AL	33.55	-86.82	71	2161	18.9	991	0.20
2	Birmingham 2, AL	33.50	-86.92	71	2161	17.4	987	0.19
3	Davie, FL	26.08	-80.24	70	2111	8.3	672	0.27
4	Pompano Beach, FL	26.22	-80.13	66	1982	8.3	713	0.24
5	Jacksonville 1, FL	30.14	-81.63	58	1712	10.3	751	0.19
6	Jacksonville 2, FL	30.36	-81.55	49	1435	10.6	612	0.23
7	Tampa 1, FL	27.93	-82.51	69	2076	11.5	753	0.21
8	Tampa 2, FL	28.05	-82.38	47	1415	11.4	552	0.20
9	Miami, FL	25.79	-80.21	67	2025	9.7	635	0.21
10	Miami, FL	25.47	-80.48	58	1765	7.8	613	0.29
11	Orlando, FL	28.55	-81.35	68	2047	10.3	762	0.21
12	Winter Park, FL	28.60	-81.36	67	1997	10.2	734	0.21
13	Belle Glade, FL	26.72	-80.67	54	1645	7.7	741	0.21
14	Royal Palm Beach, FL	26.73	-80.23	71	2161	7.9	727	0.24
15	Delray Beach, FL	26.46	-80.09	54	1645	7.4	618	0.23
16	Saint Petersburg, FL	27.79	-82.74	71	2140	10.6	728	0.19
17	Decatur, GA	33.69	-84.29	71	2152	15.9	989	0.22
18	Doraville, GA	33.90	-84.28	71	2156	16.6	979	0.23
19	Atlanta, GA	33.82	-84.39	71	2145	16.7	978	0.22
20	Louisville 1, KT	38.23	-85.82	62	1875	16.3	815	0.20

Table A.1 (Continued)

Station No.	City, State	Lat.	Lon.	Months	ALL	PM2.5	SAT days	AOT
21	Louisville 2, KT	38.19	-85.78	71	2150	16.2	950	0.20
22	Durham, NC	35.99	-78.90	71	2159	14.1	1151	0.20
23	Winston-Salem, NC	36.11	-80.23	68	2061	15.1	1071	0.21
24	Greensboro, NC	36.11	-79.80	48	1461	13.8	797	0.19
25	Charlotte 1, NC	35.22	-80.88	71	2161	15.6	1127	0.21
26	Charlotte 2, NC	35.24	-80.79	71	2161	14.9	1127	0.21
27	Raleigh, NC	35.86	-78.57	68	2069	14.0	1143	0.20
28	North Charleston, SC	32.98	-80.07	71	2161	12.4	1162	0.21
29	Charleston, SC	32.79	-79.96	70	2131	11.9	1152	0.22
30	Taylors, SC	34.90	-82.31	71	2161	14.9	1210	0.19
31	Columbia, SC	33.99	-81.02	48	1461	14.0	792	0.19
32	WEST VIEW, SC	34.93	-82.00	71	2161	14.3	1156	0.19
33	Nashville 1, TN	36.18	-86.74	71	2161	15.2	993	0.20
34	Nashville 2, TN	36.12	-86.87	71	2161	13.3	1007	0.19
35	Knoxville 1, TN	35.97	-83.95	71	2161	16.9	1091	0.22
36	Knoxville 2, TN	36.02	-83.87	71	2161	16.2	1082	0.22
37	Memphis 1, TN	35.18	-89.93	71	2158	13.9	1070	0.20
38	Memphis 2, TN	35.21	-90.03	71	2156	14.3	1061	0.21

Lat: Latitude, Lon: Longitude, ALL is number of days of data available between 08/2000 and 12/2005, PM2.5is the mean PM2.5 mass concentration (μgm^{-3}) for all days, SAT is the number of days when Terra MODIS AOT is available, and AOT is mean aerosol optical thickness at 550 nm for SAT.

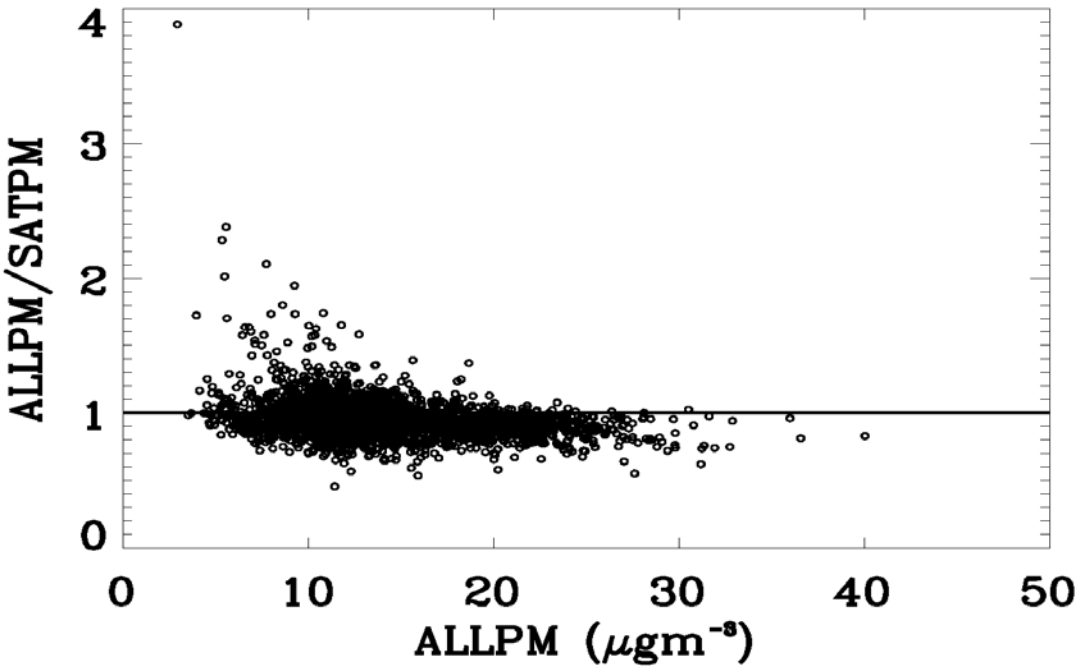

Figure A.2 Scatter plot of the ratio of the monthly mean ground PM2.5 mass concentration from all measurements (ALLPM) to the monthly mean ground PM2.5 mass concentration from only when satellite retrieval of AOT is available (SATPM) as a function of monthly mean value of ALLPM. Data from all 38 stations and 71 months is shown.

Figure A.3 shows the difference between ALLPM and SATPM for each station averaged for the entire six year period. The differences in mean values over all 71 months for each station are connected by the solid line and the gray shaded area represents the standard deviations. Also shown for each station is the percent number of days that data is available (SATDAYS). Recall that ALLPM denotes all the values averaged for the entire six year period from ground measurements, and SATPM are also values obtained from the ground measurements only when satellite retrievals were available. Several conclusions can be gleaned from Figure A.3. First, note that some stations have only 32 % data availability over a 71 month period whereas the maximum data availability is only 57%. The lack of data availability is largely due to cloud contamination and the mean SATPM is higher than ALLPM over all the stations except one, indicating that the PM2.5 values obtained during the times when satellite data is available is larger. This suggests that if we aggregate the PM2.5 values when satellite-retrievals are available, it overestimates the PM2.5 by nearly 2μgm^{-3}. This may be due to missing low PM2.5 values and sampling the higher PM2.5 values when satellite data is available. Other factors such as cloud cover, rainfall and secondary organics production may also be responsible for these differences that are difficult to unravel with only the data sets used in this study. In contrast, the ALLPM values have nearly a 100% data sampling rate, but have a high proportion of small PM2.5 values that reduces the mean values below the SATPM values. The inset of Figure A.3 presents the frequency distribution of ALLPM-SATPM and it shows that nearly 72% of the values have higher SATPM when compared to ALLPM and the remaining 28% have lower SATPM than ALLPM. The differences are between 0.0 to -2.5μgm^{-3} for 57% of the data and between 0.0 to 2.5μgm^{-3} for about

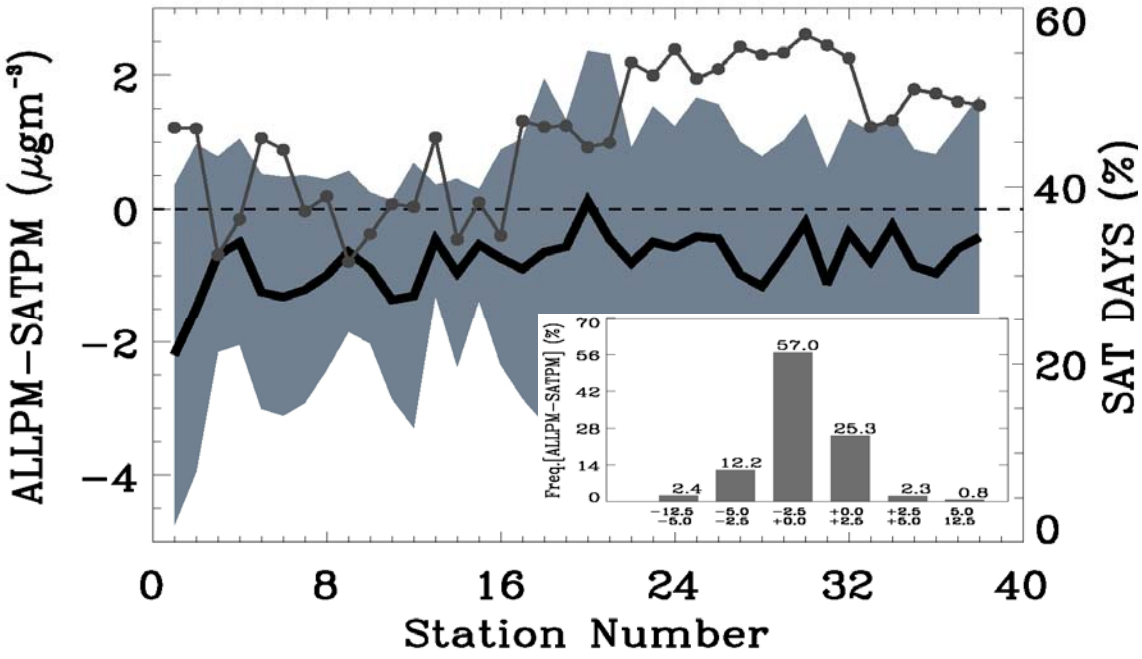

Figure A.3 Difference between PM2.5 mass from all ground-based measurements (ALLPM) and only when MODIS reports cloud-free conditions (SATPM) for 38 locations. Also shown is the data availability for each station in percent. The inset shows the frequency distribution of the difference between ALLPM and SATPM. The shaded area shows the standard deviation in difference.

25% of the data. Only less than 18% data points show greater (>±2.5) differences. Our analysis therefore indicates when averaged over monthly and yearly time scales, the surface (ALLPM) and satellite (SATPM) do not show large differences even though the satellite samples these locations less than 50% of the time. We therefore conclude that the satellite data sampling does not pose major problems for capturing PM2.5 air quality. However, we are not saying that the satellite data is a perfect surrogate for monitoring PM2.5 air quality, since it also depends upon other factors such as conversion of columnar satellite to surface values, cloud cover, relative humidity and satellite retrieval issues. Our analysis only indicates that assuming satellite is indeed a good surrogate for PM2.5 air quality, the sampling from satellites due to cloud cover and other issues does not present a major problem for studying particulate matter air quality over monthly and annual scales.

We next examine the ALLPM-SATPM differences as a function of various months, seasons and years averaged over all stations, which is a good indicator of a large spatial area analysis (Table A.2 and Figure A.4). The differences range from -2.8 to 4.9 μgm^{-3} with mean value of -0.8±1.1 μgm^{-3}. The upper range (4.9 μgm^{-3}) of the difference corresponds to February 2000 when Terra-MODIS satellite started providing data and only 12% of data were available in this initial phase. The positive differences occurred in the winter and spring months, when SATDAYS are the highest (>50%). Analysis for each month (averaged over all years) separately shows that July has a maximum difference of 2.0 μgm^{-3}, whereas January shows minimum difference of -0.07 μgm^{-3}. In terms of available SAT DAYS, November has maximum values (55%) and July has minimum values (39%). ALLPM values are smaller than SATPM values for most months

Table A.2 Annual mean statistics from 2000-2005 for ALLPM, SATPM and ALLPM-SATPM denoted as ALL-SAT.

YEAR	Parameter	MIN	MAX	MEAN	STD	No of Station	Total Days
2000	***ALLPM***	9.38	22.12	15.32	3.30	31	11169
	SATPM	9.56	25.10	16.28	3.67	31	4362
	ALL-SAT	-4.87	1.05	-1.25	1.11	31	
2001	***ALLPM***	7.66	19.01	13.73	3.39	31	11132
	SATPM	9.12	22.02	14.42	3.24	31	5376
	ALL-SAT	-8.28	1.62	-1.23	1.91	31	
2002	***ALLPM***	6.94	17.38	12.49	3.15	37	13323
	SATPM	8.09	19.51	13.68	3.27	37	5933
	ALL-SAT	-3.68	0.57	-1.73	0.95	37	
2003	***ALLPM***	7.24	17.33	12.14	3.01	37	13441
	SATPM	8.01	19.87	12.88	3.15	37	5691
	ALL-SAT	-2.78	0.86	-1.04	0.89	37	
2004	***ALLPM***	7.51	17.68	12.60	2.87	36	13110
	SATPM	8.03	20.16	13.22	2.73	36	5794
	ALL-SAT	-4.36	1.20	-0.94	1.18	36	
2005	***ALLPM***	7.21	19.67	12.74	3.33	35	12701
	SATPM	7.79	22.60	13.91	3.72	35	6055
	ALL-SAT	-4.91	0.24	-1.72	1.16	35	

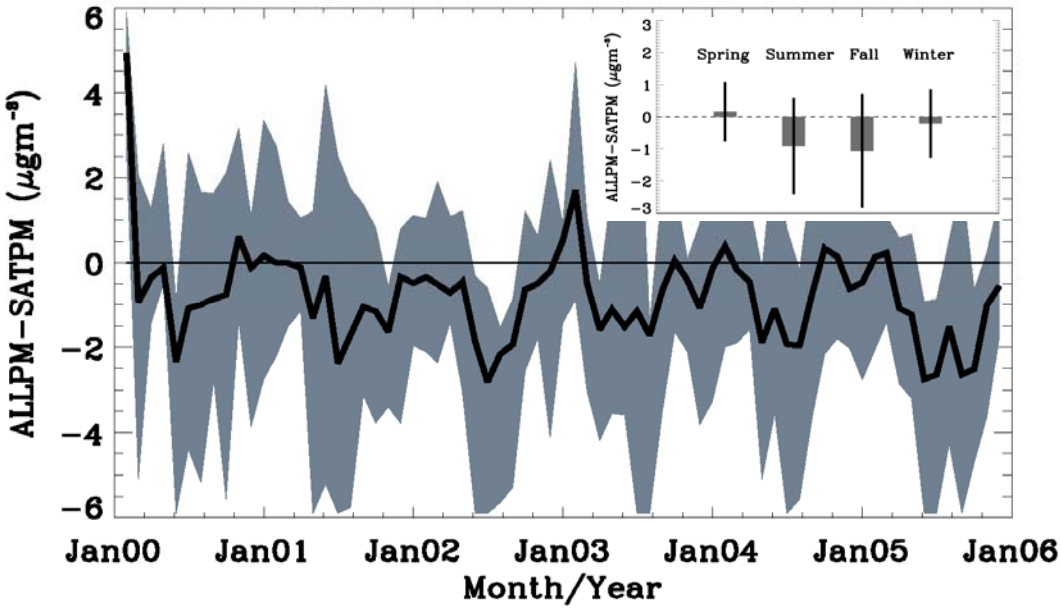

Figure A.4 Same as Figure A.2 except that the data is shown for all stations as a function of each month. The inset shows the ALLPM-SATMP differences as a function of seasons and the standard deviations are shown is vertical lines. Shaded area shows the standard deviation in the difference.

except February when ALLPM are higher by 1.1μgm^{-3} than SATPM. Going from monthly to seasonal analysis, the maximum differences occur during summer (-0.91 μgm^{-3}) and fall (-1.1 μgm^{-3}) when PM2.5 values are generally larger in this area [*e.g Wang and Christopher,* 2003]. Terra-MODIS day time cloud fraction values are also high during summer and fall month in the entire study area, thereby reducing SAT DAYS and producing these large differences. Differences in winter values are negative but very small (-0.21 μgm^{-3}) compared to fall and summer seasons, which is due to higher number of SAT DAYS in this season. The spring season shows SATPM values lower than ALLPM values with positive difference of 0.16 μgm^{-3}. Monthly and seasonal variations are also dependent on the variability in PM2.5 mass concentration within the month and season. Months with high variability in PM2.5 will tend to show large differences compared to those with low variability. Averaged over all stations, the mean ALLPM – SATPM differences are on the order of 2 μgm^{-3} indicating that satellite data does not have major sampling issues averaged over all stations. However, there may be large day to day variations in PM2.5 that cannot be captured by satellites if there is cloud-cover.

Note that it is difficult to extrapolate these results to a global context without having PM2.5 data from the ground since the differences between the ALLPM and SATPM depend upon various factors including meteorology, pollution sources, transport, vertical distribution of aerosols, clouds and other factors. For example it is possible that in a highly polluted area the PM2.5 values from the ground may be high but due to persistent cloud cover, the satellite will not have AOT retrieval. Furthermore, regions with high day to variability in PM2.5 mass will tend to produce larger differences

compared to areas with less variability. Therefore it is difficult to estimate this sampling bias for global areas without further research.

A.4 Summary and Conclusions

Polar orbiting satellites increasingly are being used for studying surface PM2.5 air pollution. The typical strategy in most studies is to develop a regression relationship between hourly or daily PM2.5 mass concentration from the ground stations and coincident satellite-derived AOT. These relationships can then be applied to larger spatial scales for determining air quality indices that range from good to unhealthy categories. While there are obvious advantages and disadvantages when using satellite data to estimate PM2.5 as outlined in this paper, one of the fundamental limitations of satellite data is the unavailability of air pollution observations when clouds obstruct the satellite sensors field of view. This poses the question then as to how well do the satellites represent PM2.5 air quality if a location is not being sampled due to cloud cover and other reasons such as high reflectivity surfaces such as urban areas and when snow/ice conditions prevail. To examine this issue, we used six years of data over 38 locations in Southeastern United States and aggregated all PM2.5 values from the ground (ALLPM) over monthly, seasonal and yearly time scales. For these stations, we used 5X5 boxes grid cells centered on the PM2.5 location and obtained PM2.5 values from the ground when the satellite retrieval of AOT is available (SATPM). The difference between ALLPM and SATPM provides a measure of the sampling issue when satellites are used. We reiterate that we are not focusing on the robustness of columnar AOT to obtain surface PM2.5 mass that has been addressed in previous studies. We merely address the

issue of whether satellite data can adequately sample the PM2.5 over monthly to yearly time scales. Our results indicate that over monthly and yearly time scales, whether for individual stations or area averages, the difference is between the ALLPM and SATPM is less than 2 μgm^{-3} indicating that that loss of data due to non retrieval conditions such as cloud cover, and surface type not a major problem. However, we note that these results cannot be extrapolated to global areas without further research.

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