**Atmospheric
Chemistry
and Physics
Discussions**

Particulate matter air
quality

P. Gupta and
S. A. Christopher

Title Page

Abstract	Introduction
Conclusions	References
Tables	Figures

|◄◄| |►►|

|◄| |►|

| Back | Close |

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

EGU

Seven year particulate matter air quality assessment from surface and satellite measurements

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Abstract

Using seven years of the Moderate Resolution Imaging Spectroradiometer (MODIS) aerosol optical thickness (AOT) data and ground measurements of particulate matter mass over one site in the Southeastern United States (33.55 N, 86.82 W) we present a comprehensive analysis of various aspects of particulate matter air quality. Monthly, seasonal and inter-annual relationships are examined with emphasis on sampling biases, quality indicators in the AOT product and various cloud clearing criteria. Our results indicate that $PM_{2.5}$ mass concentration over Northern Birmingham has decreased by about 23% in year 2006 when compared to year 2002 and air quality during summer months are poor when compared to winter months. MODIS-Terra AOT data was available only about 50% of the time due to cloud cover and favorable surface conditions. However, the mean difference in monthly mean $PM_{2.5}$ was less than $2.2\,\mu gm^{-3}$ derived using all the data and from only those days when satellite AOT was available indicating that satellite data does not have sampling issues. The correlation between $PM_{2.5}$ and MODIS AOT increased from 0.52 to 0.62 when hourly $PM_{2.5}$ data were used instead of daily mean $PM_{2.5}$ data. Changing box size for satellite data around the ground station during comparisons produced less than ±0.03 difference in mean AOT values for 90% of observations. Application of AOT quality flags reduced the sample size but does not affect AOT-$PM_{2.5}$ relationship significantly. We recommend using AOT quality flags for daily analysis, whereas long time scale analysis can be performed without using all AOT retrievals to obtain better sampling. Our analysis indicates that satellite data is a useful tool for monitoring particulate matter air quality especially in regions where ground measurements are not available.

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,

EGU

Particulate matter air quality

P. Gupta and
S. A. Christopher

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
5 from surface based stations.

Advancement in satellite remote sensing of aerosols 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
10 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 2.5 μm
15 ($PM_{2.5}$). Most of these studies have compared MODIS derived AOT with surface measured $PM_{2.5}$ mass concentration and linear regression equations were formulated to calculate $PM_{2.5}$ 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
20 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 $PM_{2.5}$ mass concentration (Donkelaar et al., 2006).

Satellite remote sensing studies indicate that AOT data can be used to monitor $PM_{2.5}$
25 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 are 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 $PM_{2.5}$ mass concentration over North Birmingham (NBHM) to assess particulate matter quality from daily to yearly scales using various methods. We focus on a single location (NBHM) and provide detailed air quality analysis while considering different aspects of surface and satellite measurements over a seven year time period. Previous studies in this region or elsewhere used data for limited time periods and primarily focused on estimation of relationship between $PM_{2.5}$ and AOT only. In addition to the $PM_{2.5}$-AOT relationship, we also examine the differences in surface and satellite observations on monthly and yearly time scales. This provides insights into the effects of aerosol quality flags and on data loss, and we finally provide recommendations on using satellite data to derive particulate matter air quality.

Particulate matter air quality

P. Gupta and
S. A. Christopher

Title Page

Abstract   Introduction

Conclusions   References

Tables   Figures

◄◄   ►►

◄   ►

Back   Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

EGU

2 Data

The data sets used in current study can be categorized into satellite and surface observations. The first is the Terra-MODIS AOT data product and the second is daily and 1 h mean PM$_{2.5}$ mass concentration obtained from NBHM. Both data sets are obtained
5 for a seven-year time period (2000–2006).

2.1 MODIS

MODIS Level 2 aerosol data (MOD04, Collection 5) from 24 February 2000 to 30 June 2006 were obtained from the Atmosphere Archive and Distribution System (LAADS) at NASA's Goddard Space Flight Center (GSFC). Each MOD04 granule contains the
10 aerosol properties both over land and ocean retrieved from 5 min 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, b). The parameters used are the spectral aerosol optical thickness (AOT), Angstrom exponent (AE), Cloud fraction (CF), and quality flags related to AOT retrieval, cloud masking,
15 and surface types (Table 1). 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 PM$_{2.5}$ measurements, nine different criteria are applied to evaluate the
20 AOT product for PM$_{2.5}$ air quality applications (Table 2). To understand these criteria, a brief discussion of MODIS collection 5 aerosol retrieval over land 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 2.1 μm channel along with the vegetation
25 index to obtain surface reflectance (Levy et al., 2007a) and other channels to mask cloudy pixels. Although the retrieved AOT values are reported at a 10×10 km^2 pixel resolution, the algorithm initially uses radiance measurements at much higher resolution

331

of $0.5{\times}0.5\,\text{km}^2$. All 400 pixels (20×20 pixels), each having resolution of $0.5{\times}0.5\,\text{km}^2$ within the $10{\times}10\,\text{km}^2$ area, are first examined for cloud contamination (Martin et al., 2002). The MODIS internal cloud mask (Ackerman et al., 1998) at $1{\times}1\,\text{km}^2$ spatial resolution is also used to identify cirrus. All pixels identified as cloudy are removed

5 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 $10{\times}10\,\text{km}^2$ pixel. Levy et al. (2007a) provide further details.

10 The percentage of cloudy pixels in a $10{\times}10\,\text{km}^2$ grid is computed using $1{\times}1\,\text{km}^2$ 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, 2007). In this study, we have used quality control flags

15 listed in criterion 4 through 9 (Table 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 $PM_{2.5}$ measurements. This flag contains 40 bits of information, which includes confi-

20 dence 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), which indicates

25 confidence level of AOT retrieval at two wavelengths.

EGU

2.2 Particulate matter mass (PM$_{2.5}$)

Particulate matter mass concentration (μgm^{-3}) with aerodynamic diameter less than 2.5 μm (PM$_{2.5}$) is regularly monitored by United States Environment Protection Agency (USEPA). The particulate matter air quality in the Continental United States is evaluated
5 based on the PM$_{2.5}$ mass concentration measured using a network of surface stations. Generally, a Tapered Element Oscillating Microbalance (TEOM) instrument is used to measure the mass of PM$_{2.5}$ 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
10 particulate air pollutants near the surface. PM$_{2.5}$ data from these networks include 24 h average (daily) concentration data and continuous (hourly) PM$_{2.5}$ mass concentration measurements. We use PM$_{2.5}$ data from the North Birmingham (NBHM, 33.55 N and 86.82 W) station located in Jefferson County, Alabama, which has a data availability of 85% for any given year. Figure 1 shows a detailed view of the stations along with larger
15 view of the EPA region 4.

3 Methodology

To obtain coincident PM$_{2.5}$ and MODIS AOT, nine different criteria are applied (Table 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
20 10×10 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 5×5 pixel box over
25 the surface measurement locations and 15-min observations over one hour to repre-sent a similar air mass as observed by MODIS. This was justified by examining the

Particulate matter air quality

P. Gupta and
S. A. Christopher

normal speed of aerosol transport (50 km/h) in the mid-troposphere 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).

5   In the current study, three different box sizes (Fig. 1) centered on NBHM are evaluated to verify the assumptions used in Ichoku et al. (2002). Table 2 provides the nine criteria used to obtain MODIS AOT. Criteria numbers 1 to 3 represents box sizes of 5×5, 4×4, and 3×3 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
10   applying any quality flags on MODIS AOT since this is the standard practice in most studies (e.g. Wang and Christopher, 2003).

Criteria numbers 4 to 9 use the 3×3 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 sta-
15   tions are retrieved over land surface with solar zenith angles less than 60°. In some instances there could be inland water pixels or highly reflecting surfaces that can create uncertainties in AOT values (Hutchinson et al., 2005).

Criteria numbers 5 to 7 include the quality flags associated with cloud cover (CMQA) in the $10×10$ km^2 MODIS pixel. Cloud mask quality flags are associated with percent-
20   age area of $10×10$ 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,
25   while at the same time the quality of the AOT value increases. However, reducing the number of pixels may also introduce sampling bias in the mean AOT value.

Criteria number 8 corresponds to confidence levels in AOT retrieval, which is based on number of pixels (at 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

Particulate matter air quality

P. Gupta and
S. A. Christopher

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). Criteria number 9 applies all of the restrictions of criteria number 8 plus, it allows only those pixels for which cloud cover is less than 30%. 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 $PM_{2.5}$ mass concentration to evaluate the long term changes in AOT values as well as $PM_{2.5}$ mass concentrations.

4 Results

Results are divided into 4 different sections. Section one discusses the multi-year trends in $PM_{2.5}$ mass concentration, $PM_{2.5}$ 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 $PM_{2.5}$ 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 $PM_{2.5}$ mass concentration on hourly, daily, monthly, and annual basis. This section also evaluates the stability of AOT-$PM_{2.5}$ relationships during different seasons and years. The final section, which is unique to the current study, is evaluation of MODIS AOT for $PM_{2.5}$ research by considering sampling issues as well as quality flags associated with each retrieved AOT pixel. This section also revisits the results of Sect. 1 to 3 under these new criteria for obtaining satellite observations and discusses the differences in AOT and $PM_{2.5}$ that arise when using stringent quality flags.

P. Gupta and
S. A. Christopher

EGU

4.1 Long term air quality trends over North Birmingham

PM$_{2.5}$ mass concentrations 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 qual-
5   ity. Figure 2 shows the time series of 24-h mean PM$_{2.5}$ mass concentration along with monthly and annual mean trends. Daily variations in PM$_{2.5}$ mass are represented by thin light gray line, which are mainly associated with local meteorological conditions and changes in local emissions of PM$_{2.5}$ particles. High values could also be associated with transport of pollution from surrounding areas. The background colors denote the
10   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 35 μgm^{-3} are the annual and 24 h mean national standards set by USEPA under National Ambient Air Quality Standards (NAAQS). The previous (prior to 17 December 2006) 24 h standard of 65 μgm^{-3} is also plotted for
15   comparison purposes (Fig. 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 PM$_{2.5}$ mass concentration of 75.3 μgm^{-3} was observed on 25 October 2000 whereas the average of all daily mean values is 18.7±9.7 μgm^{-3}. The thick gray line presents the
20   monthly mean PM$_{2.5}$ mass concentration calculated using daily mean values. The monthly mean values clearly show a seasonal trend with high values during 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
25   particle production. Similar conditions do not occur in the winter months, thereby lim-iting PM$_{2.5}$ production. Seasonal changes in local meteorology also affect the PM$_{2.5}$ production and removal processes. The month-to-month variation is not the same for all the years and varies slightly with peak values around July–August. Monthly

mean $PM_{2.5}$ mass peaks at $34.6\,\mu gm^{-3}$ during October 2000 with a minimum value of $11.3\,\mu gm^{-3}$ observed in January 2003. A weak decreasing trend in annual $PM_{2.5}$ was noted with the highest and lowest $PM_{2.5}$ mass concentration of $22.1\,\mu gm^{-3}$ and $17.0\,\mu gm^{-3}$ observed in 2002 and 2006, respectively.

Figure 3 presents similar analysis for MODIS AOT using criteria 1 from Table 2. Recall that this criterion includes all AOT data without any quality flags. The AOT shows similar trends as $PM_{2.5}$ mass with high values during summer and low values during winter months. Seasonal changes in AOT are more prominent compared to seasonal changes in $PM_{2.5}$ mass concentration due to sensitivity of urban aerosols towards relative humidity (Wang et al., 2007). The absolute difference in $PM_{2.5}$ mass from winter to summer is not as high (when compared to $PM_{2.5}$ trends in Fig. 2) when compared to MODIS AOT. Recall that AOT is an optical property which is function of light scattering from the aerosol particle and it increases in size in summer time due to particle growth under high relative humidity 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, $PM_{2.5}$ mass does not change significantly due to growth of particles as TEOM make measurement of dry particles under about 40% relative humidity conditions.

Figure 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 7 years time and are 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. 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

EGU

group (USG) 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 $PM_{2.5}$ concentration.

Day to day variations in $PM_{2.5}$ mass concentrations is usually associated with variations in local meteorological conditions. Meteorological conditions that strongly control $PM_{2.5}$ 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. High levels of pollution can be observed during strong temperature inversions near surface (Kukkonen et al., 2005) and the magnitude, vertical structure, and temporal evolution of inversions significantly influence the air quality (Hussein et al., 2006).

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 $PM_{2.5}$ mass concentration over NBHM". To answer this question, we assume that $PM_{2.5}$ mass can be estimated using MODIS AOT data. To test this assumption, daily mean $PM_{2.5}$ measurements are collocated with MODIS AOT data according to criteria 1 as given in Table 2. Although the Terra satellite overpasses NBHM site almost every day around 10:30 a.m. local time, AOT retrieval may not be available on a daily basis due to clouds and other limitations (e.g. bright

Particulate matter air quality

P. Gupta and
S. A. Christopher

EGU

surface during certain seasons). Figure 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
5 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.
10 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 5b presents the monthly mean difference in $PM_{2.5}$ mass concentration between all 24-h mean $PM_{2.5}$ measurements averaged for each month (ALL) and $PM_{2.5}$
15 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 $PM_{2.5}$ mass. Negative differences indicate that using $PM_{2.5}$ data during the time of the satellite overpass overestimates $PM_{2.5}$ on monthly as well as yearly basis. This indicates that events with low
20 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 are during the winter season. The mean difference is $-2.2\,\mu\mathrm{gm}^{-3}$ with a maximum negative value of $-9.7\,\mu\mathrm{gm}^{-3}$ during May 2004. The maximum positive difference of
25 $6.1\,\mu\mathrm{gm}^{-3}$ was obtained during February 2000 since Terra-MODIS started observations on 24 February 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 $-0.5\,\mu\mathrm{gm}^{-3}$ whereas February has a positive difference of $0.6\,\mu\mathrm{gm}^{-3}$ Again, summer months have large negative differences

EGU

compared to winter months, which corresponds to more number of available days in winter and less during summer months (Table 3). The solid line with dots in Fig. 4b shows the yearly mean values, which are always negative. The maximum difference $(-3.0\,\mu\mathrm{gm}^{-3})$ in annual mean was in 2001 whereas minimum difference $(-1.2\,\mu\mathrm{gm}^{-3})$ was during 2006.

4.3 Regression analysis between PM$_{2.5}$ and MODIS AOT

Particulate matter air quality monitoring from satellite is based on the relationship between satellite derived AOT and surface measured PM$_{2.5}$ mass concentration (Wang and Christopher, 2003; Gupta et al., 2006). The accuracy of satellite estimated PM$_{2.5}$ over any given location depends on the robustness of the regression relationship between AOT and PM$_{2.5}$ 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 PM$_{2.5}$ mass concentration and hence their inclusion in the AOT-PM$_{2.5}$ relationship can improve the overall accuracies in PM$_{2.5}$ mass estimation from satellite observations (Gupta et al., 2006). However, we do not attempt to add meteorological parameters to the analysis for the current paper. A separate manuscript is under preparation, which accounts for meteorological conditions and other ancillary data sets.

Computation of a single-variate least-squares linear regression relationship between MODIS AOT and PM$_{2.5}$ 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 PM$_{2.5}$ are explored. Figure 6a presents the month to month and annual variations of MODIS AOT (primary y-axis) along with daily mean PM$_{2.5}$ mass concentration (secondary y-axis). Both MODIS AOT and PM$_{2.5}$ mass concentration follow similar trends on monthly and yearly time scales as noted in Sect. 4.1. Figure 5b shows hourly averaged (red lines) PM$_{2.5}$ during Terra MODIS overpass time from 2000 to 2006 along with 24 h averaged PM$_{2.5}$ (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 24 h averages to mean obtained using hourly average $PM_{2.5}$ is 1.66±0.47.

The linear correlation coefficient between 24 h mean $PM_{2.5}$ mass concentration and MODIS AOT is 0.52 with the regression relationship given by Eq. (1).

$$PM_{2.5} = 15.8 + 27.5 \times \text{MODIS AOT} \tag{1}$$

When a similar analysis is performed on hourly averaged $PM_{2.5}$ data, the correlation increases to 0.62 and the corresponding regression equation becomes Eq. (2).

$$PM_{2.5} = 8.8 + 29.4 \times \text{MODIS AOT} \tag{2}$$

These regression equations can be used to estimate the ground level $PM_{2.5}$ mass concentration. As expected, the instantaneous MODIS AOT correlate better with hourly averaged $PM_{2.5}$ compared to 24 h averaged $PM_{2.5}$ due to diurnal variations in $PM_{2.5}$ mass measurements. National Air Quality Standards are set for 24 h mean $PM_{2.5}$ mass concentration and it is an important parameter to monitor, hence further analysis is focused only on 24 h mean $PM_{2.5}$ and despite its moderate correlation, these relationship can estimate daily air quality categories with a high degree of accuracy (Wang and Christopher et al., 2003).

Several studies have shown that the relationship between $PM_{2.5}$ 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 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

EGU

less than 0.1 over land (Remer et al., 2005; Levy et al., 2007a). These low AOT's correspond to low $PM_{2.5}$ concentrations and in general, summer and fall correlations are higher compared to winter and spring. Highest correlations are associated with mean high AOT and $PM_{2.5}$ mass concentration, which is consistent with our previous
5   analysis in other parts of the world (Gupta et al., 2006; 2007).

Intercept in regression equation represents the background levels of $PM_{2.5}$ 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 dif-
10   ficult. 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 $PM_{2.5}$ mass in respective months. The slope of this relationship is an important parameter for converting AOT into surface $PM_{2.5}$ mass.
15   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
20   relationships because same dry $PM_{2.5}$ mass concentration under different relative humidity condition can produce different AOT due to changes in scattering properties of aerosols.

4.4   MODIS AOT, box size, and quality flags

Satellite aerosol retrieval algorithms convert the measured reflectance to a geophysical
25   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 MODIS AOT product. The MODIS science team recommends using these quality flags along with actual AOT values for research studies. Depending on

Particulate matter air quality

P. Gupta and
S. A. Christopher

Title Page

Abstract | Introduction

Conclusions | References

Tables | Figures

◄◄ | ►►

◄ | ►

Back | Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

EGU

the application, 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
5 values is quantitatively estimated for three different box sizes. Table 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.

10 4.4.1   Varying box size around ground station

Criteria numbers 1 to 3 (CN1 to CN3) in Table 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 5×5 (0.5×0.5°, AOT5), 4×4 (0.4×0.4°, AOT4), and 3×3 (0.3×0.3°, AOT3) pixels. Decreasing box size, decreases the number of pixels used to derive MODIS
15 AOT values for NBHM, and may affect the overall mean AOT and these can change $PM_{2.5}$ 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 compared to small box sizes, which can overestimate or under estimate AOT values compared to point measurements from ground stations. Ichoku et
20 al. (2002) reported that a 5×5 box is a good approximation for AOT validation with 1 h averaged AERONET AOT values, which is supported by arguing that average speed of aerosol transport in mid-troposphere is about 50 km/h 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
25 measured at the surface in the form of $PM_{2.5}$ mass concentration. Since the average travel speed of aerosols (air mass) near the surface (lowest few hundred meters) is much less ($<4\,ms^{-1}$) than mid-troposphere, one hour averaged $PM_{2.5}$ measurements may not represent the air mass in the 50×50 km area sampled by MODIS instrument.

As a result, a smaller 3×3 pixel 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 7 presents differences in daily, monthly, and annual mean AOTs due to changes in box sizes. The inset in Fig. 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 on 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 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 (AOT5) 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 (0.05±0.15×AOT) 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 4). It is also important to note that, reducing box size from 5×5 to 3×3 reduces the number of available data by about 8%. The regression coefficients for the AOT-PM$_{2.5}$ relationship (see Sect. 4.3) 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%.

4.4.2 Applying quality flags

Criteria numbers 3 to 9 (CN3 to CN9) in Table 2 use a constant box size of 3×3 pixels around the PM$_{2.5}$ 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, 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 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 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 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 arise due to sampling issues discussed in the previous section. Table 6 presents these differences for each month separately to shows seasonal variability in AOT for different criteria. Overall, there is not much of a 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 9 presents the reanalysis of regression equations as discussed in Sect. 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. 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 $PM_{2.5}$ 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 $PM_{2.5}$ and AOT does not vary significantly and only improves from 0.52 to 0.55 from CN1 to CN9.

5   Summary

Seven year surface $PM_{2.5}$ 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 $PM_{2.5}$ mass concentration and satellite derived integrated columnar aerosol loading (AOT) with the new generation of Collection 5 MODIS aerosol products. The application of quality control flags on aerosol optical thickness values and effects on these flags on the $PM_{2.5}$-AOT relationships were also examined.

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 $2\,\mu\mathrm{gm}^{-3}$ when compared to using a 100% data availability rate from ground measurements. Our analysis of quality control flags and box sizes for evaluating $PM_{2.5}$-AOT

EGU

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 $PM_{2.5}$-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.

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P. Gupta and
S. A. Christopher

EGU

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Particulate matter air quality

P. Gupta and
S. A. Christopher

Title Page

Abstract	Introduction
Conclusions	References
Tables	Figures

◄◄ ►►

◄ ►

Back	Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

EGU

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Particulate matter air quality

P. Gupta and
S. A. Christopher

Title Page

Abstract | Introduction

Conclusions | References

Tables | Figures

◄ | ►

◄ | ►

Back | Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

EGU

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Particulate matter air quality

P. Gupta and
S. A. Christopher

Title Page

Abstract | Introduction

Conclusions | References

Tables | Figures

|◄ | ►|

◄ | ►

Back | Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

EGU

Table 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 $0.55\,\mu m$ for both ocean (best) and land (corrected)
5	Cloud_Fraction_Land	Cloud cover fraction for each pixel determined by spatial technique (Martin et al., 2002)
6	Cloud_Mask_QA	Cloud mask quality assurance originally derived at 1×1 km that is recomputed at 10×10 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 f,or AOT over land contains product quality flags, retrieval processing flags and input data resource flags. Stores in 5 byte SDS.

Particulate matter air quality

P. Gupta and
S. A. Christopher

Table 2. Criteria used to obtain coincident MODIS–Terra AOT data over PM$_{2.5}$ locations.

Criteria No.	Box size around ground stations	Surface type	SZA (deg)	AOT Flag	Cloud Flag
		Quality Flags			
1	±0.25 deg. ($\sim$5×5 pixels)	–	–	–	–
2	±0.20 deg. ($\sim$4×4 pixels)	–	–	–	–
3	±0.15 deg. ($\sim$3×3 pixels)	–	–	–	–
4	±0.15 deg. ($\sim$3×3 pixels)	Land	≤60	–	–
5	±0.15 deg. ($\sim$3×3 pixels)	Land	≤60	–	0–90 %
6	±0.15 deg. ($\sim$3×3 pixels)	Land	≤60	–	0–60 %
7	±0.15 deg. ($\sim$3×3 pixels)	Land	≤60	–	0–30 %
8	±0.15 deg. ($\sim$3×3 pixels)	Land	≤60	G and VG	–
9	±0.15 deg. ($\sim$3×3 pixels)	Land	≤60	G and VG	0–30%

SZA: Solar Zenith Angle, AOT: Aerosol Optical Thickness.

Table 3. Regression coefficients, mean and standard deviation (σ) of $PM_{2.5}$ and MODIS AOT.

	Slope	Intercept	R	Counts	$PM_{2.5}$	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	9.59	0.20	0.16

P. Gupta and
S. A. Christopher

Title Page

Abstract | Introduction

Conclusions | References

Tables | Figures

|◀ | ▶|

◀ | ▶

Back | Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

EGU

Table 4. Effect of box size on AOT values at daily, monthly and yearly scales.

Box	Min.	Max.	Daily 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

Table 5. Effect of quality flags on mean AOT values over the stations. Daily, monthly and yearly differences (BEST-AOT).

Difference	Daily 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

EGU

Table 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

EGU

EGU
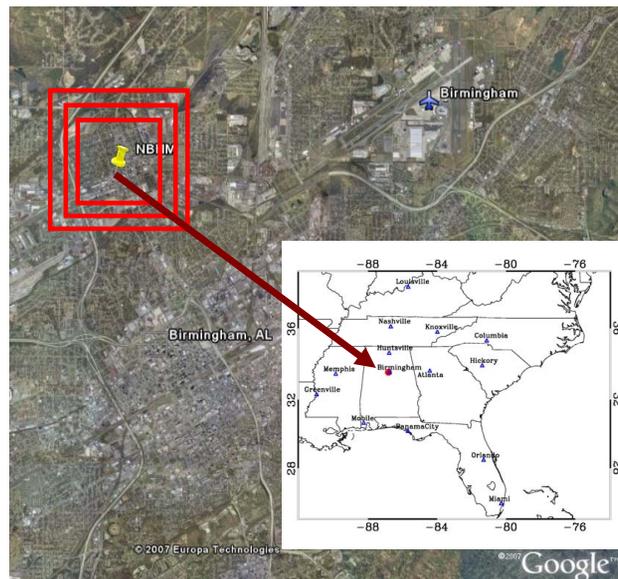
Fig. 1. Study area with location of PM$_{2.5}$ ground stations in Birmingham, AL. Also shown is a zoomed satellite image (courtesy Google) of the region with three different box sizes (3×3, 4×4, and 5×5) used to collocate MODIS observations. Red colored boxes are just relative representation of box size and not drawn on actual pixel size scale.
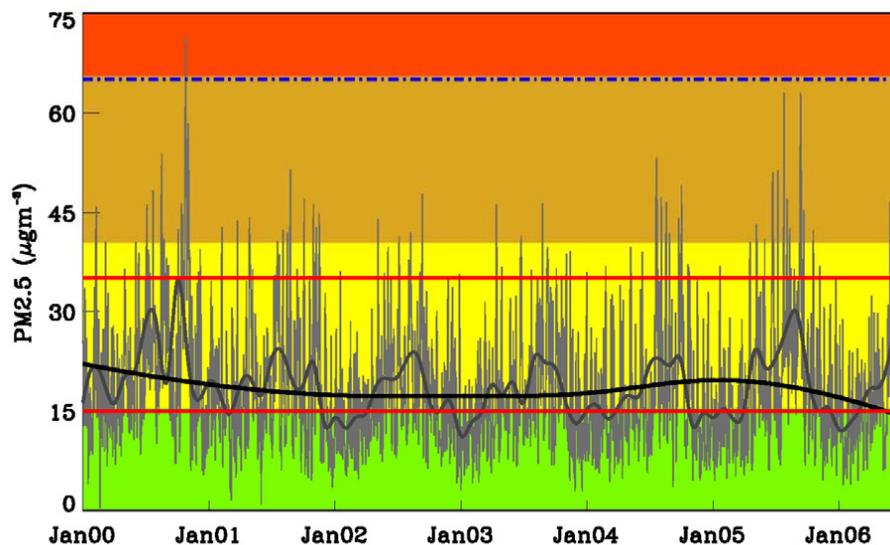
Fig. 2. Time series analysis of PM$_{2.5}$ mass concentration over NBHM site starting from January 2000 to June 2006. Background colors show air quality categories. Daily variations are shown in thin light gray line, monthly mean in thick gray line and yearly means are shown in thick black line. Two red lines show new daily and annual national standard for PM$_{2.5}$ mass whereas dotted blue line shows old annual USEPA standards.
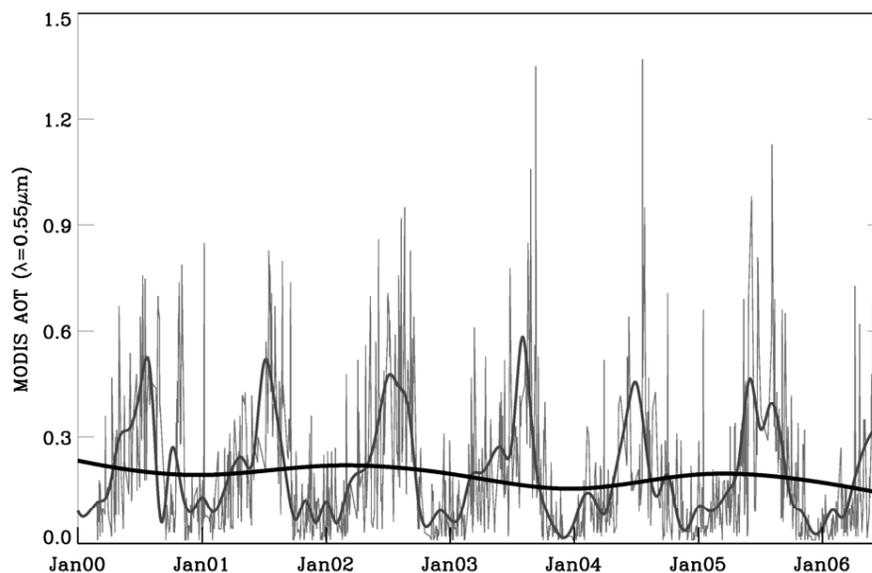
Fig. 3. Time series analysis of MODIS AOT over NBHM from 24 February 2000 to June 2006. Daily variations are represented by thin light gray line, monthly mean represented by thick gray line and yearly means are shown as thick black line.
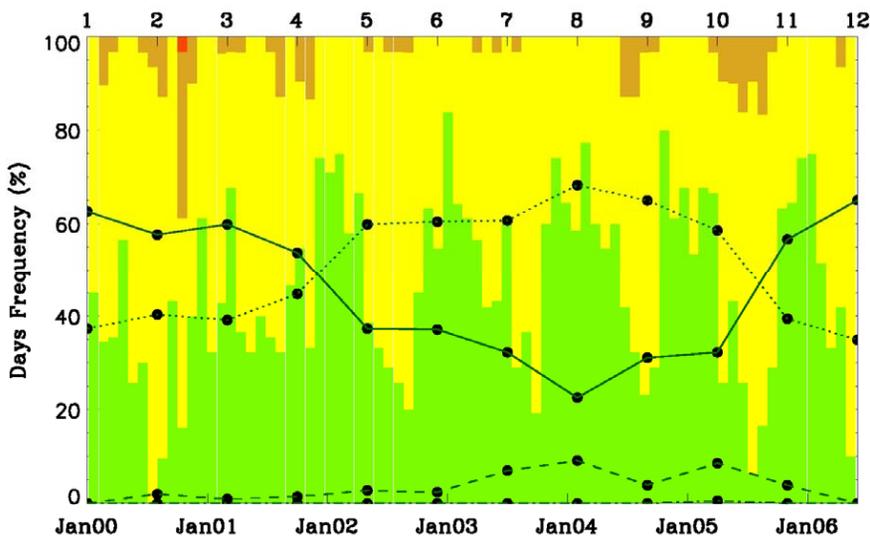
Fig. 4. Frequency distribution of number of days (%) falling in any particular air quality category during each month over NBHM starting from February 2000 to June 2006. Air quality categories are derived from PM$_{2.5}$ mass concentration as defined by USEPA and represented by different colors. Green 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.

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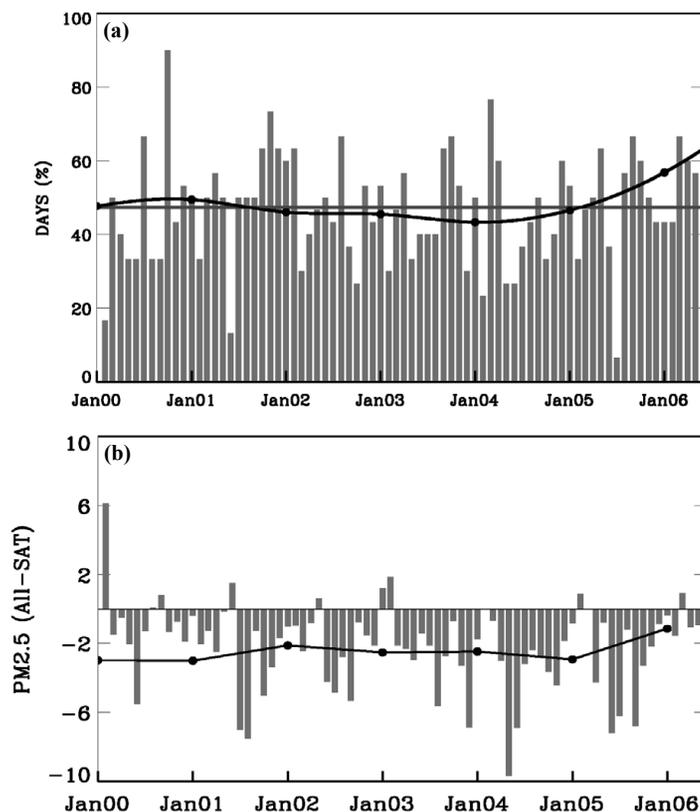
Fig. 5. Time series analysis of difference in PM$_{2.5}$ mass over NBHM site starting from February 2000 to June 2006. **(a)** number of days (%) MODIS AOT data are available over NBHAM and **(b)** the monthly mean PM$_{2.5}$ mass from all ground observation minus PM$_{2.5}$ mass from only those days 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 47.4%.

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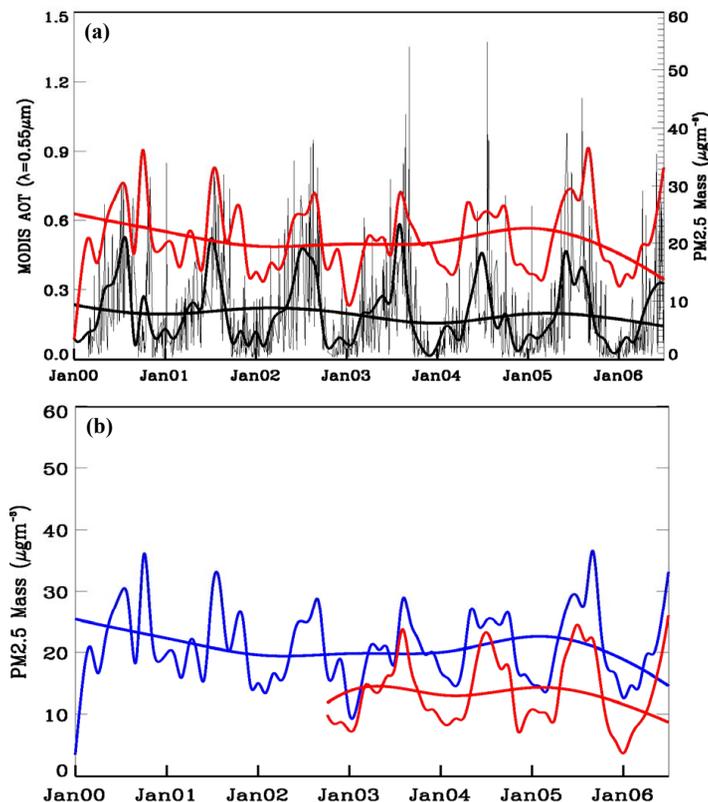
Fig. 6. Trend analysis of surface measured PM$_{2.5}$ mass (1 h and 24 h) and MODIS AOT over the station. **(a)** daily, monthly and annual mean AOT as time series (black lines) and monthly and annual mean PM$_{2.5}$ mass concentration (red lines). **(b)** monthly and annual mean PM$_{2.5}$ mass obtained from 24 h mean PM$_{2.5}$ mass data (blue lines) along with monthly and annual mean PM$_{2.5}$ mass obtained from 1 h (during satellite overpass) PM$_{2.5}$ data (red lines).
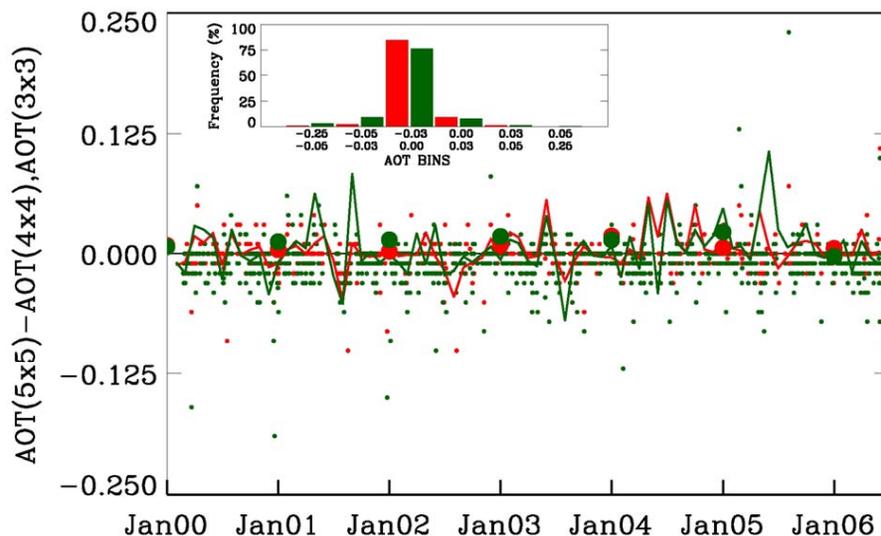
Fig. 7. Time series analysis of difference in AOTs due to change in box size around the ground station. Y-axis represents AOT5-AOT4 (red color in plot) and AOT5-AOT3 (green color in plot). Big filled circles are yearly mean, solid lines are monthly mean and small filled circles are daily AOT values. The inset panel shows frequency distribution of difference in AOTs for daily values.

Title Page

Abstract | Introduction

Conclusions | References

Tables | Figures

◄◄ | ►►

◄ | ►

Back | Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

EGU
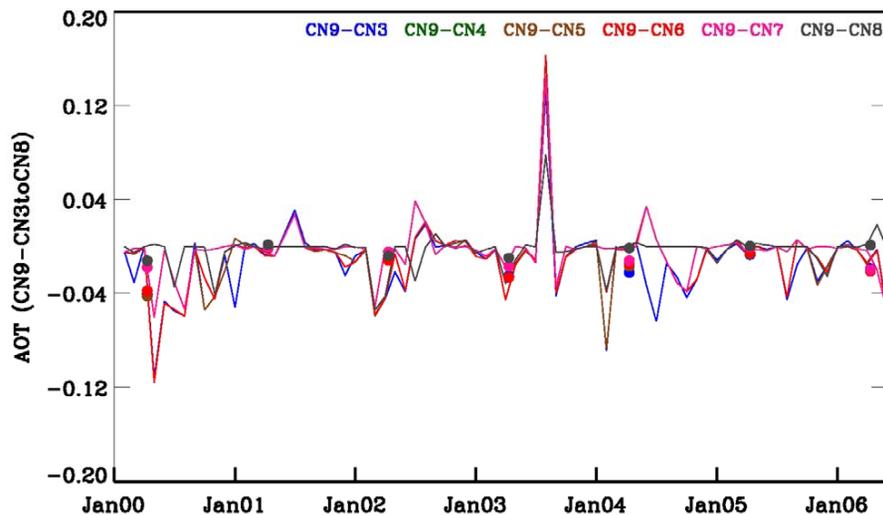
Fig. 8. Time series analysis of difference in AOTs after applying quality flags. Y-axis represents Best AOT (CN9) – all other AOTs (CN3 to CN8). Big filled circles are yearly mean, solid lines are monthly means.
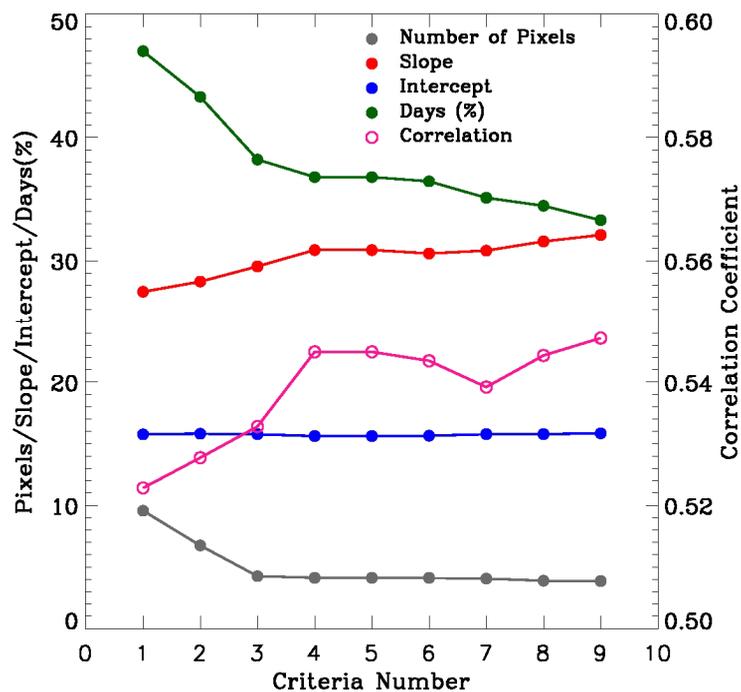
Fig. 9. Revisiting regression analysis using AOT from nine different criteria.

Title Page

Abstract | Introduction

Conclusions | References

Tables | Figures

◄ | ►

◄ | ►

Back | Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

EGU