Reduction of Aerosol Absorption in Beijing since 2007 from MODIS and AERONET

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Abstract. An analysis of the time series of MODIS-based and AERONET aerosol records over Beijing reveals two distinct periods, before and after 2007. The MODIS data from both the Terra and Aqua satellites were processed with the new Multi-Angle Implementation of Atmospheric Correction (MAIAC) algorithm. A comparison of MAIAC and AERONET AOT shows that whereas MAIAC consistently underestimated peak AOT values by 10-20% in the prior period, the bias mostly disappears after mid-2007. Independent analysis of the AERONET dataset reveals little or no change in the effective radii of the fine and coarse fractions and of the Angstrom exponent. At the same time, it shows an increasing trend in the single scattering albedo, by ~0.02 in 9 years. As MAIAC was using the same aerosol model for the entire 2000-2010 period, the decrease in AOT

bias after 2007 can be explained only by a corresponding decrease of aerosol absorption caused by a reduction in local black carbon emissions. The observed changes correlate in time with the Chinese government's broad measures to improve air quality in Beijing during preparations for the Summer Olympics of 2008.

1. Introduction

Anthropogenic aerosols play an important climate role directly, by scattering and absorbing solar radiation, and indirectly by modifying cloud properties. Due to the complexity of aerosol processes and incomplete understanding of their interactions with the climate system, large uncertainties in climate modeling remain [*IPCC*, 2007]. The role of aerosols can change dramatically from cooling to warming the ambient atmosphere with an increase in their absorption, which is modulated by the amount of black carbon (BC). Anthropogenic BC is generated mainly from fossil-fuel combustion and biomass burning; the first is found to be twice as efficient a warming agent as the second [*Ramana et al*., 2010].

Space-based remote sensing has been used to study the aerosol optical thickness (AOT) of Earth's atmosphere. Currently, the global operational AOT data over land are provided by MODIS [*Levy et al*., 2007; *Hsu et al.*, 2004], MISR [*Martonchik et al*., 2009; *Kahn et al*., 2010], and, in the case of absorbing aerosols, OMI [*Torres et al.*, 2007]. Available space-based remote sensing has limited ability to evaluate aerosol absorption. MISR can distinguish two-to-four categories of absorption under favorable retrieval conditions [*Chen et al*., 2008], whereas OMI can detect aerosols that are absorbing in the UV (Dust and organic/black carbon). The MODIS Dark Target algorithm (MOD04) retrieves AOT and Fine Mode Fraction (FMF) based on a regionally prescribed scattering and absorbing properties of the fine and coarse components derived from Aerosol Robotic Network of Sun photometer (AERONET) [*Holben et al*., 1998] data.

Beijing is a growing megacity with a population in 2007 of over 17 million people. The very high level of air pollution in Beijing has multiple consequences, including reduced visibility and human health impacts [e.g., *Lei et al*., 2010]. Industry, residential heating and cooking, and the transportation sectors are the main local aerosol emission sources [*Lei et al*., 2010]. In the past decade, the Chinese government undertook measures to improve air quality, starting with the 2001 Law of Air Pollution and Control, and moving aggressively since 2005 as part of preparations for the 2008 Summer Olympics. A recession that began in the 3rd quarter of 2008 and lasted through 2009 provided a continued follow-on effect [*Lin et al*., 2010]. Data analysis showed the effectiveness of the government measures in decreasing SO_2 during the pre-Olympic period [*Guinot et al*., 2007]. Although a reduction of BC was reported [*Wang et al*., 2005], its assessments remain uncertain; for example, *Guinot et al*. [2007] found that with the decrease in total carbon (TC), the BC/TC ratio in the fine aerosol mode has increased from 21% to 28% over the period 2000-2004. The present article provides new information on aerosol absorption trends in Beijing, based on an analysis of AERONET data, and MODIS data processed by the new Multi-Angle Implementation of Atmospheric Correction (MAIAC) algorithm [*Lyapustin et al*., 2010].

2. Methods and Results

MAIAC is a new algorithm developed for MODIS, which retrieves AOT and FMF over land simultaneously with surface bidirectional reflectance parameters derived from time series analysis and image-based processing. Retrievals are performed at 1 km resolution. Following the MOD04 algorithm, models for the fine and coarse aerosol fractions are specified regionally. A comparison between MAIAC and MOD04 shows that these algorithms have generally similar accuracy over vegetated or relatively dark surfaces, and that MAIAC improves accuracy over brighter surfaces [*Lyapustin et al*., 2010], which include most urban areas.

In this work, we consider MAIAC aerosol retrievals over Beijing (116.381°E, 39.977°N) using 50×50 km^2 subsets from MODIS centered on selected AERONET sites. The subsets are provided by the MODIS Operational Adaptive Processing System (MODAPS) for both the Terra and Aqua satellites. A comparison with AERONET was performed, following the MODIS validation strategy [*Ichoku et al*., 2002]: a) The AERONET level 2 measurements are averaged over ±30 min intervals centered on overpass time. A comparison is considered valid if there are at least two measurements within the 1-hour interval. b) MAIAC data are discarded if cloud fraction in the averaging area exceeds 20% or if snow is detected.

The 1 km MAIAC AOT data show high aerosol spatial variability in the Beijing area. To account for this factor, two averaging window sizes, 10 and 20 km, were compared (50 km windows are used in MOD04 validation). The smaller window, which was found to improve correlation (r^2) by ~0.01-0.02 and to provide better matches with AERONET at peak AOT values, was used in this work. A similar result over urban areas was reported in the MISR validation study [*Kahn et al*., 2010].

Figure 1 shows a time series of AERONET and MAIAC AOT at 0.47 µm for MODIS Terra (top) and Aqua (bottom). Visual inspection indicates two distinct periods for both Terra and Aqua: prior to ~March 2007 (period I), MAIAC rather consistently underestimates AOT by 10-20% at peak values. In the subsequent period (II), this bias mostly disappears. To verify this observation, separate MAIAC-AERONET AOT regression plots (not shown) were generated for periods I and II. Both Terra and Aqua show an increase in the regression slope, from 0.94 to 0.99 for Terra, and from 0.91 to 1.0 for Aqua between periods I and II, respectively, at average r^2=0.88-0.92. The regression slope is strongly controlled by the aerosol absorption. Increasing the imaginary part of refractive index n_i by 0.001 (n_i=0.009, single scattering albedo SSA=0.90) brings the regression slope to 1.02 for Terra and to 0.99 for Aqua for period I.

Figure 2 shows a time series for the AOT difference $\Delta = AOT_{AER} - AOT_{MAIAC}$ for MODIS Terra (left) and Aqua (right). These data represent all cases of moderate-to-high AOT selected according to the AERONET data (AOT_{AER}>0.4). For MODIS Terra, we filtered out six outliers with higher difference values ±(0.6-0.8). Removing these (symmetric) outliers enhances coherence of the plot and has a negligible effect on the trend lines. Both Terra and Aqua show the reduction of MAIAC's bias with respect to AERONET by approximately 0.1 (Aqua) and 0.06 (Terra) over 9 years. Separating periods I and II, Aqua shows a smaller trend, and Terra shows no trend, during period I. In period II, both datasets show larger, and similar, negative trends. Reducing the threshold AOT_{AER} from 0.4 to 0.3 does not change the observed trends. The smaller change for Terra than for Aqua may be related to the diurnal cycle of SSA in Beijing [*Garland et al.*, 2009].

MAIAC retrievals for the entire time series were performed using the same bi-lognormal size distribution with volumetric radius (R_v) and standard deviation (σ) of the fine (F) and coarse (C) fractions: R_v^F =0.15+0.05τ, R_v^F <0.25 µm, σ^F =0.45 µm; R_v^C =2.5+0.2τ, R_v^C <3 µm, σ^C =0.6 µm, where τ is aerosol optical thickness at 0.47 µm. The dynamic growth of aerosol size with optical thickness reflects experimentally established hygroscopic particle growth [*Meier et al.*, 2009]. The complex refractive index used in the visible spectrum (n_r=1.44; n_i=0.008) gives SSA=0.91 at 0.47 µm for a typical summertime Angstrom parameter 1.2-1.3 [*Eck et al.*, 2010]. Since the same aerosol model was used in MAIAC processing for the entire period 2000-2010, the observed difference between periods I and II is most easily explained by a change in aerosol particle size and/or absorption over time, e.g., due to reduction in BC emissions. At large AOT values ~1.5-3 typical for Beijing, it is absorption that mainly controls sensitivity of retrieved AOT, which can be as high as 10-15% in response to a small SSA change of 0.01.

Next, we analyzed the AERONET inversion products [*Dubovik and King*, 2000; *Dubovik et al*., 2002] (effective radii (R_{ef}) of the fine and coarse fractions and SSA) and Ångström parameter (*b*). Springtime dust events [*Eck et al*., 2010] were excluded, based on the condition $b<0.6$. To ensure adequate accuracy of the inversion results, the AERONET Level 2 inversion quality criteria were used (≥21 azimuthal angles; solar zenith angle ≥50 °; 5% residual error; $AOT_{0.44}>0.4$ for SSA) [*Holben et al*., 2006; *Dubovik et al*., 2002; *Smirnov et al*., 2000]. We analyzed both daily and monthly averages. Both datasets give the same results. The monthly averages are plotted in Figure 3, which shows no significant change in R_{ef}^{F}. The coarse mode radius was found to increase less than retrieval uncertainties, from 2.2 to 2.3 μm, which does not affect our findings. The Ångström parameter, which is an integral indicator of size and relative concentration for the fine and coarse aerosol fractions, shows no trend. On the other hand, the SSA was found to increase by ~0.02 over 9 years at the 88% confidence level, which confirms the above MAIAC-AERONET analysis. To exclude effects of Beijing aerosol absorption seasonality [*Eck et al*., 2010] and uneven sampling, the SSA analysis was repeated with the winter months excluded. This removed most values below 0.85, but gave a similar result for the SSA increase (0.017), bringing it closer to the MAIAC assessment (~0.01). Finally, to understand the spatial extent of the effect, two sites in addition to Beijing were analyzed - XiangHe, located about 60km southeast of Beijing in the downwind direction, and Taihu (120.215°E, 31.421°N) located about 1000 km south of Beijing. Both sites have shorter time records, starting in summer 2005. As expected, results for XiangHe show an increase in SSA (~0.03) similar to nearby Beijing, whereas data from 2005 till June 2010 for the remote Taihu site show a very small negative trend.

3. Discussion

A few years prior to the Beijing Olympic Games, the Chinese government embarked on efforts to lower air pollution in the Olympic host cities, including Beijing and its five surrounding provinces, targeting primarily the significant pollution sources such as transportation, industrial, and coal-burning activities [e.g., *Wang et al.*, 2010]. As a result, over 50,000 coal-burning homes were converted to natural gas. A number of industrial polluters were either modernized, including 16,000 coal-burning factories, or shut down by 2008. Many factories were relocated from Beijing, including the 4th largest steel company (Shougang Group), whose production was cut by 73% in 2008, and was completely phased out in December 2010. After 2007, the diesel vehicle emission standard was raised from Euro III to Euro IV in Beijing and from Euro II to Euro III for the rest of the country. Although a reduction in total air pollution in Beijing since 2005 [*Lin et al.*, 2010] and during the 2008 Olympics [*Cermak and Knutti*, 2009] as a result of these measures has been reported, there are several mitigating factors: *a*) although emissions of regulated sources such as SO_2 and primary particulate matter (PM) from cement production, coal combustion, and biofuel burning, were reduced, emissions of other less-controlled aerosol precursors (e.g., nitrogen compounds) are estimated to have grown [*Lin et al.*, 2010]; *b*) GDP has increased over 9% annually since 2007 except in 2009 (~6%), and the number of vehicles in Beijing and throughout the country has grown by 49% for the period 2007-2010; *c*) the meteorology and regional aerosols in Beijing play an important role [*Streets et al.*, 2007], which complicates the analysis, as discussed below.

Regional contributions to the Beijing aerosol load vary depending on their origin: highly absorbing aerosol, due mainly to residential heating and cooking (SSA=0.8-0.85), is brought in from the rural North, whereas the heavily industrialized South-West and South brings low absorbing, secondary aerosols (SSA=0.9-0.95), whose absorption is further reduced by hygroscopic growth in moist air [*Garland et al.*, 2009; *Eck et al.*, 2010]. A particular aerosol realization is the superposition of diurnal [*Garland et al*., 2009], synoptic (~5-7 days) [*Streets et al*., 2007; *Jia et al*., 2008], and seasonal [*Eck et al*., 2010] patterns. Synoptic variation is linked to the passage of cold fronts from the North, which produce a gradual rotation of wind direction from the North to the South-West/South, with a simultaneous reduction of wind speed, creating stagnant conditions. This periodic variation gives rise to a saw-tooth-like pattern of suspended PM [*Jia et al*., 2008], which is superimposed on the local diurnal cycle. This behavior is modulated by the seasonal pattern, which is dominated by northern airmasses in winter and humid southern airmasses in summer. Source apportionment simulations [*Matsui et al*., 2009] reveal that for Beijing, the primary aerosols, including BC, are controlled by emissions from within 100 km around the city over the preceding 24 h, whereas emissions from as far away as 500 km over the previous three days were found to affect secondary aerosols.

Although regional aerosols make it difficult to assess the total PM reduction, they have less effect on aerosol absorption and its change with time. Indeed, the local aerosols contain highly absorbing BC from primary sources. The regional (secondary) aerosol may contain as much as, or more of the total carbon, but its light-absorption efficiency is significantly reduced due to the chemical/physical modifications of hygroscopic growth and aging. Even though regulations reduced total carbon, the absorption properties of secondary aerosols could not change much. By contrast, the reduction of BC from local (primary) sources leads to a measurable reduction of total absorption, as detected and described in this article. Our finding is supported by the long-term measurements of PM chemical composition in Beijing [*Okuda et al*., 2011], which reported an observed 33% BC reduction in 2008 as compared to the reference period of 2005-2007.

4. Conclusions

In this work, we demonstrated a measurable reduction of the aerosol absorption over Beijing during 2007-2010 compared to the previous five years, most probably caused by the regulation of BC emissions. Two independent methods based on the time series analysis of a) MODIS-AERONET AOT data, and b) AERONET SSA data, give similar results, which are in qualitative agreement with in situ chemical composition analysis [*Okuda et al*., 2011]. The coarse assessment based on the MAIAC-AERONET AOT regression slopes indicates a reduction of the imaginary refractive index of about 0.001, corresponding to an increase in single scattering albedo by ~0.01, close to the AERONET SSA-change assessment of ~0.02. The timing of these changes is correlated with expansive measures adopted by the Chinese government to improve air quality in Beijing in the wake of the 2008 Olympic Games.

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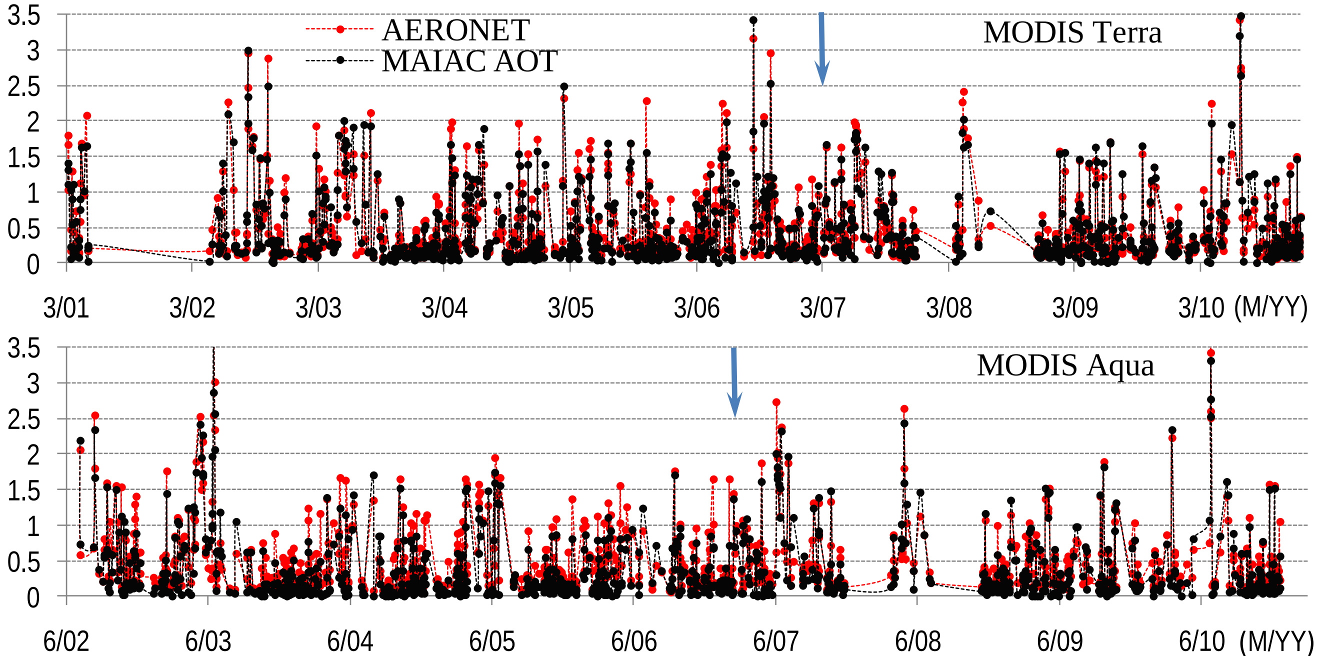
Figure 1. Time series showing MAIAC and AERONET AOT data over Beijing at 0.47 µm, for 10 years of MODIS Terra (top) and ~9 years of MODIS Aqua (bottom). The arrow indicates the approximate boundary between periods I and II.

Figure 2. Time series difference plots (AERONET - MAIAC AOT)$_{0.47}$ for moderate-to-high atmospheric opacity (AERONET AOT>0.4) over Beijing, showing MODIS Terra (left) and Aqua (right).

Figure 3. Time series plots of AERONET effective radius for fine aerosol fraction (left) and single scattering albedo (right) for the Beijing site. Points represent monthly averaged values.



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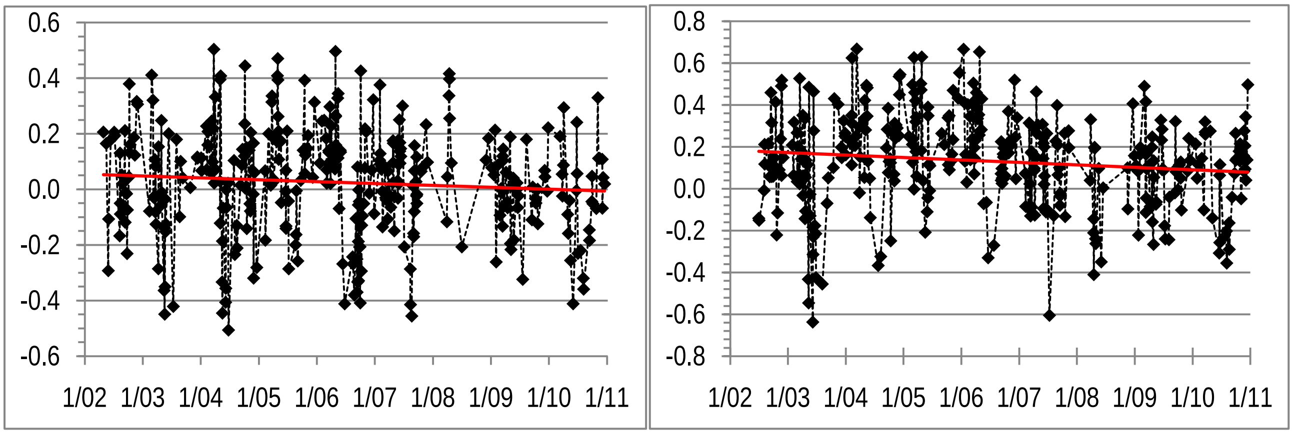

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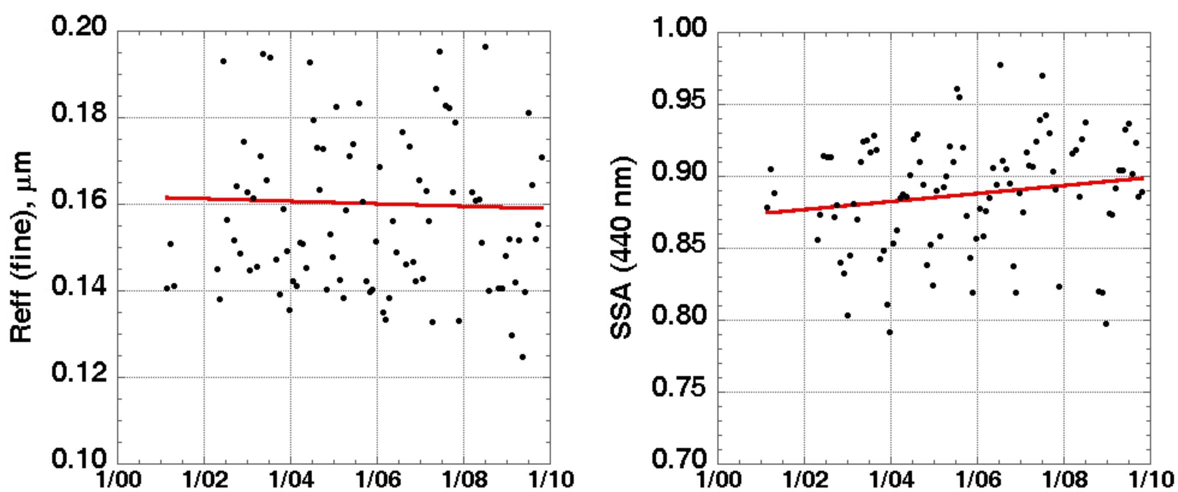

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