Correlation Analysis between AOD and Cloud Parameters to Study Their Relationship over China Using MODIS Data (2003–2013): Impact on Cloud Formation and Climate Change

Na Kang, Kanike Raghavendra Kumar*, Yan Yin, Yiwei Diao, Xingna Yu

Key Laboratory for Aerosol-Cloud-Precipitation of China Meteorological Administration, School of Atmospheric Physics, Nanjing University of Information Science and Technology, Nanjing 210044, Jiangsu, China

ABSTRACT

In the present study, we examined the spatial and temporal variations in aerosol optical depth (AOD) at 550 nm and its relationship with various cloud parameters derived from the Moderate resolution Imaging Spectroradiometer (MODIS) sensor onboard Terra satellite. The data have been analyzed for the period of 10-years between March 2003 and February 2013 over 12 major cities in China. The results revealed that high AOD noticed over low latitude regions influenced with high anthropogenic activities and the low AOD observed for the high altitude and mountainous areas since AOD accounts for the slant path which reduces the aerosol emissions. Also the aerosol variations in the atmosphere are complicated by several factors in emissions (natural and anthropogenic) as well as stagnant synoptic meteorology. From the temporal studies, it is clear that the maximum AOD found during summer followed by spring and autumn with a minimum AOD in winter season for all the regions of study in China. Further, we studied the relationship between AOD versus water vapor (WV), cloud fraction (CF), cloud optical thickness (COT), cloud effective radius (CER), cloud top pressure (CTP), and cloud top temperature (CTT) for the selected regions in China. Also, we performed regression analysis and one paired student’s t-Test to represent the probability of data significant at 95% confidence for the derived AOD values and cloud parameters in order to provide a better understanding of aerosol-cloud interaction.

Keywords: MODIS; China; Aerosol optical depth; Cloud parameters; Correlation Coefficient.

INTRODUCTION

Atmospheric aerosols produced from different natural and man-made activities are mixed together and hence each aerosol particle is a composite of different chemical constituents, which determine the refractive index of aerosols (Ranjan et al., 2007; Alam et al., 2010). Aerosols affect the Earth’s climate in many characteristics ways (Li, 2004; Tai et al., 2010; Lee and Penner, 2011). They can affect the energy balance of the earth-atmosphere system by direct scattering and absorption of radiation (Satheesh and Moorthy, 2005; Wright et al., 2010). Aerosols are known to impact the formation and the life cycle of clouds. A wide range of measurements have shown that anthropogenic aerosols can change clouds and their optical properties (Ackerman et al., 2000; Rosenfeld, 2000; Kim et al., 2003; Koren et al., 2004; Alam et al., 2010; Balakrishnaiah et al., 2012; Kumar, 2013; Alam et al. 2014). It is important to understand and quantify the microphysical impact of both natural and anthropogenic aerosols on clouds, in order to understand and predict climate change (Forest et al., 2002; Kuntti et al., 2002; Anderson et al., 2003).

Aerosols have many direct, semi-direct and indirect effects on clouds. The indirect effects, or aerosol-cloud interactions, result from more cloud condensation nuclei (CCN) creating a dense of smaller particles for a given amount of cloud water. This makes the clouds brighter (first indirect effect or Twomey effect; Twomey, 1977; Kaufman and Fraser, 1997; Feingold et al., 2003), as well as affecting the resulting lifetime of the clouds in complex ways (second indirect effect, or lifetime effect; Albrecht, 1989; Quaas et al., 2008, 2010). Both the first and second indirect effects act to cool the atmosphere, partially offsetting warming due to greenhouse gases (Lohmann and Feichter, 2005). However, absorbing aerosols such as soot emitted from biomass burning can suppress cloud formation by warming the atmosphere, increasing evaporation of water droplets and also increasing the atmospheric stability, which is known as the semi-direct effect (Hansen et al., 1997; Ackerman et al., 2000; Johnson et al., 2004).

The aerosol-cloud interactions play a significant role in
global climate; however, there are large uncertainties in the magnitude of the forcing (IPCC, 2013). Therefore, aerosols, clouds, and their interaction with climate is still the most distinct area of climate change. To assess the regional and global climate change caused by aerosols on clouds, detailed information is required on the atmospheric concentrations of aerosol in the region (Dutkiewicz et al., 2009). A number of studies on aerosol-cloud interactions have been conducted in the south and east parts of the Chinese continent to address the spatial and temporal variability of aerosols and clouds (e.g., Alam et al., 2010; Balakrishnaiah et al., 2012; Kumar, 2013, 2014; Alam et al., 2014; Tang et al., 2014). More details on the literature have been reviewed to understand the climatic impact of aerosols on cloud properties and are described in the supplementary material (SM, S1).

Many investigations on the aerosol optical depth (AOD) data have been conducted to study the pollution over China since the launch of Moderate Resolution Imaging Spectroradiometer (MODIS) satellite (Li et al., 2007; Chan, 2009; He et al., 2012; Dong et al., 2013; Luo et al., 2013). To the best of our knowledge, only a few studies have focused on the relationship between AOD and cloud properties over China (e.g., Tang et al., 2014). The present study investigates the spatial and temporal variations of aerosol properties over 12 metro cities in China based on the 10-year long-term (March 2003–February 2013) data derived from MODIS onboard Terra satellite. Further, investigations have been carried out to study the impact of aerosol on cloud parameters such as water vapor (WV), cloud fraction (CF), cloud optical thickness (COT), cloud effective radius (CER), cloud top pressure (CTP), and cloud top temperature (CTT).

DATA AND METHODOLOGY

MODIS Satellite

The MODIS instruments allow the scientists to study many of the Earth’s terrestrial and oceanic characteristics with a single instrument. The MODIS is a remote sensor with two Earth Observing System (EOS) Terra and Aqua satellites which provide an opportunity to study aerosols from space with high accuracy (Xiong et al., 2009). The daytime Aqua overpass (13:30 local solar time (LST)) is chosen over the Terra overpass (10:30 LST) since clouds are more likely to be developed in the afternoon than in the morning. The MODIS instrument provides high radiometric sensitivity (12 bit) in 36 spectral bands, ranging in wavelength from 0.41 µm to 14.4 µm. Out of 36 spectral bands, two are imaged at a nominal spatial resolution of 250 m at nadir, with five bands at 500 m, and the remaining 29 bands at 1 km.

MODIS has supplied useful information on aerosols, clouds, moisture, and the surface since its launch on the Terra satellite in December 1999 (King et al., 1992). MODIS products are becoming important data sources for studies on the physical characteristics and radiative forcing of aerosols, and global climate change. MODIS uses visible and near-infrared bands to determine optical and microphysical cloud properties (Remer et al., 2005; Levy et al., 2007). The data are useful for collecting various statistics on aerosol concentration and the impacts that aerosols have on cloud formation (Kumar, 2013; Alam et al., 2014). For water vapor, the retrieval for the near-infrared region during clear sky condition is adopted. More detailed information on algorithms for the retrieval of aerosol and cloud parameters is available at http://modis-atmos.gsfc.nasa.gov.

Study Regions and Satellite Data

The present study has been carried out over 12 major cities located in the mainland of China which serves as the Provinces capitals. The geographical map and terrain of China is shown in Fig. 1 denotes the locations of the 12 selected regions with red solid circles. The regions used in this study, namely Changchun (North China, CC), Beijing (North China Plain, BJ), Hohhot (Inner Mongolia Province, HH), Xining (Northwest China, XN), Urumqi (West China, UQ), Nanjing (East China or Yangtze River Delta region, NJ), Wuhan (Central China, WH), Chengdu (Sichuan Basin, CD), Lhasa (Tibet Plateau region, LH), Guangzhou (Southeast China or Pearl River Delta region, GZ), Nanning (South China, NN) and Kunming (Southwest China, KM). Table 1 lists the information about the sites in detail. This study provides an opportunity to understand spatial and temporal variations of MODIS derived AOD and also investigated the relationship between AOD$_{550}$ and cloud parameters to assess the regional climate change caused by aerosols on clouds.

The MODIS datasets used in this study were downloaded from NASA GIOVANNI website (http://disc.sci.gsfc.nasa.gov/giovanni) of Level-3 Collection 5.1 for the 10-year period from March 2003 to February 2013. Level-3 MODIS data were quality checked and globally gridded over $1^\circ \times 1^\circ$ grid spatial resolution. The obtained atmosphere monthly products of aerosol and cloud parameters data from MODIS Terra sensor were averaged into the seasonal and annual values. In the present study, we focused on the AOD at a wavelength of 550 nm over land, as this is close to the peak of the solar spectrum and is therefore associated with major radiative effects. The AOD data used in the present study were retrieved for clear and partially cloudy skies during the period of the study whereas, we used WV values for the clear sky days between 2003 and 2013 derived from the MODIS (see SM, S2). Expected error over land in MODIS AOD was characterized as $\Delta \tau = \pm 0.05 \pm 0.15\tau$, where $\tau$ is AOD at 550 nm (Levy et al., 2007). The MODIS provides data for various cloud parameters for daytime and nighttime, either separately or combined and the daytime data only has been used for the present study.

Statistical Method

The statistical methods used in the present study were based on covariances or linear correlations. Since the variables were not normally distributed, the bivariate correlation between the variables was performed based on the Pearson and Spearman correlation analysis. The statistical analyses were performed using PASW statistics 17.0 (SPSS Inc., Chicago, IL, USA) for Windows software. The statistical significance of the difference between the AOD and cloud properties were analyzed by using one tailed distribution single paired student’s $t$-Test and analysis of
Fig. 1. (a) Geographical locations of the 12 study regions (red circles) with its provincial boundaries. (b) The terrain map of China.

Table 1. Geographical information about regions of study in China.

| S. No. | Region/Capital city | Province     | Latitude (N)  | Longitude (E) | Altitude ASL (m) | Population (~million) | Area (km²) |
|--------|---------------------|--------------|---------------|---------------|------------------|-----------------------|------------|
| 1      | Changchun (CC)      | Jilin        | 43°54′        | 125°12′       | 221              | 7.7                   | 20,532     |
| 2      | Beijing (BJ)        | Beijing City | 39°54′        | 116°24′       | 44               | 21.2                  | 16,801     |
| 3      | Hohhot (HH)         | Inner Mongolia | 40°49′       | 111°39′       | 1,065            | 2.9                   | 17,224     |
| 4      | Xining (XN)         | Qinghai      | 36°38′        | 101°46′       | 2,275            | 2.2                   | 7,372      |
| 5      | Urumqi (UQ)         | Xinjiang     | 43°49′        | 87°36′        | 800              | 3.1                   | 14,577     |
| 6      | Nanjing (NJ)        | Jiangsu      | 32°03′        | 118°46′       | 20               | 8.2                   | 6,598      |
| 7      | Wuhan (WH)          | Hubei        | 30°35′        | 114°17′       | 37               | 10.2                  | 8,494      |
| 8      | Chengdu (CD)        | Sichuan      | 30°39′        | 104°03′       | 500              | 14.1                  | 12,132     |
| 9      | Lhasa (LH)          | Tibet        | 29°39′        | 91°07′        | 3,490            | 0.6                   | 29,274     |
| 10     | Guangzhou (GZ)      | Guangdong    | 23°08′        | 113°16′       | 21               | 14.0                  | 7,434      |
| 11     | Nanning (NN)        | Guangxi      | 22°49′        | 108°19′       | 126              | 6.7                   | 22,189     |
| 12     | Kunming (KM)        | Yunnan       | 25°04′        | 102°41′       | 1,892            | 6.5                   | 21,015     |

ASL-Above Sea Level. The population provided is as per 2010 census record. Source: Wikipedia.
Spatial Distribution of AOD, AE and Water Vapor

AOD is an optical property of an aerosol column that describes the extent to which aerosol scattering or absorption will reduce solar radiation before it reaches the surface of the Earth (Guo et al., 2014). Generally, AOD is proportional to aerosol mass within the column and thus, it is used here to estimate the distribution of different aerosols. The spatial distribution patterns of AOD, AE and WV over China derived from MODIS were averaged from March 2003 to February 2013 represents the 10-year climatology over the region is shown in Fig. 2. The areas with white background in all the panels represent the missing of the data. Fig. 2(a) represents the spatial distribution of AOD at 550 nm which shows that the aerosols had a marked impact on 12 selected regions of China. These regions were chosen because most of them are major urban, industrial, suburban, coastal, desert and mountainous areas of China, with generally dense populations and due to the extreme variations in their geographic locations (see Table 1), which results in different weather patterns that, in turn, affect the aerosol load in each area (Alam et al., 2010).

High values of AOD (>0.7) were noticed over the regions in the North China Plain (BJ), East China or YRD (NJ), Central China (WH), and Sichuan Basin (CD) and moderate AOD values (0.5–0.7) in the areas of Inner Mongolia Province (HH), Northwest China (XN), Southeast China (GZ), and South China (NN). Low AOD values between 0.2 and 0.3 are observed in North China (CC), West China (UQ), Tibet Province (LH), and Southwest China (KM). The maximum AOD in the north extended through East China regions, and the very low AOD values are observed in the areas of desert dust emitted from the Taklimakan desert (Zhang et al., 2003; Luo et al., 2013) in the Tarim Basin. The low AOD was noticed over the high altitude mountainous areas (low and mid latitudes) of Tibet Province (LH) and Yunnan Province (KM) in the Southwest of China. This is due to the fact that aerosols are usually confined to within the planetary boundary layer (PBL); hence AOD decreases with altitude due to a reduced PBL height. Also it is to be considered that AOD is a columnar measurement and depends on the slant path/angle of view from a satellite. Also, the very low AOD values are observed in the areas of dense natural forest vegetation cover and sparse population in the high-latitude regions of CC, HH, XN, UQ, and KM.

The Shandong Province which is above to the Nanjing has lower urbanization compared with YRD, but it is vulnerable to dust transported from North China region. A high AOD also observed in the high latitude region of Xinjiang Province in West part of China (UQ) with prevalence of high natural aerosols dominated by desert dust emitted from the Taklimakan desert (Zhang et al., 2003; Luo et al., 2013) in the Tarim Basin. The low AOD was noticed over the high altitude mountainous areas (low and mid latitudes) of Tibet Province (LH) and Yunnan Province (KM) in the Southwest of China. This is due to the fact that aerosols are usually confined to within the planetary boundary layer (PBL); hence AOD decreases with altitude due to a reduced PBL height. Also it is to be considered that AOD is a columnar measurement and depends on the slant path/angle of view from a satellite. Also, the very low AOD values are observed in the areas of dense natural forest vegetation cover and sparse population in the high-latitude regions of CC, HH, XN, UQ, and KM.
Fig. 2. Spatial distribution of ten-year averaged (a) AOD$_{550}$ (b) Ångström exponent (AE, $\alpha_{470/660}$) and (c) Water vapor (WV) over China retrieved from MODIS Terra products for the period 2003–2013. The locations of 12 study regions are the same as that shown in Fig. 1.
scarcity of human activities and confinement of large particles due to topography contributes to high AE values (low AOD). Conversely, anthropogenic activities produce large amounts of fine-mode aerosol particles in densely populated urban and industrial areas. However, airborne dust originating from rush vehicular traffic and construction activities in urban areas, in addition to larger particle sizes of soot aerosols produced by industrial and civil coal fuel consumption (Li et al., 2003; He et al., 2012), cause coarse-mode aerosols to occupy larger proportions of aerosol particles, thus resulting in smaller AE values in urban regions than in suburban and mountainous regions.

The spatial variation of water vapor (WV) averaged for the 10-year period over China was shown in Fig. 2(c) varies...
between 0.2 and 5.0 cm. The figure reveals a good one-to-one correspondence between WV and AOD in all the regions representing strong positive correlation. From the figure it is evident that WV was found to be high (> 3.0 cm) in the southern part (coastal regions) compared to the lower values (0.2–1.3 cm) in the northern part of China with dense vegetation/forest and desert; and a moderate values ranging from 1.5 to 3.0 cm in the east and central parts of China. The box plot shown in Fig. 3(c) representing the annual mean WV showed high value of 4.3 cm in Lhasa (LH) and more or less similar values of WV over GZ and NN (3.9 cm) and NJ and CD (2.5 cm) in the southern part of China and YRD region of East China, respectively. The present analysis demonstrated that high WV will be regionally observed over locations where AE is high due to the impact WV on hygroscopic growth of aerosols which in turn increases the AOD. Kaufman et al. (2005) reported that the hygroscopic nature of aerosols mainly depends on the general synoptic meteorological conditions, which is not discussed in the present study. Xin et al. (2007) found that the high AOD values associated with the continuous anthropogenic emissions, stable meteorological conditions by weak wind speeds and slowly moving air masses are generally accompanied by higher WV.

Seasonal Variations in AOD

The inter-annual monthly mean variation in AOD$_{550}$ along with the standard deviation was shown in Table 2 for the selected regions during the period of study from 2003 to 2013. There was no data for the winter months (December–February) during the 10-year period over Urumqi (UQ) station and is due to intense snow cover and fog which restricts MODIS satellite to look into this region. It can be inferred from Table 2 that AOD shows a consistent variation with monthly low value occurring during December in most of the regions of study except for the high latitude (CC, HH, and XN) and mountainous stations (LH) where the AOD was found low in September/October. The AOD peaks during the month of June followed by May in most of the regions except for XN, UQ, CD, GZ and NN where it peaks during March/April. The standard deviations were high during the summer months, which indicate more variability in the individual AOD values during these months compared to other months. On average for the entire period of study, the high value of AOD noticed over Wuhan (0.74 ± 0.15) and a low value of 0.20 ± 0.06 at Hohhot, Xining and Kunming (see Table 2).

High (low) annual mean AOD values were observed in almost all the regions during the summer (winter) season. Data from NJ, WH, GZ, CD and BJ reveals very high mean AOD values in summer in the increasing order of 0.88 ± 0.29, 0.80 ± 0.31, 0.66 ± 0.23, 0.62 ± 0.21, and 0.57 ± 0.20, respectively during the study period (2003–2013) as compared to the other regions in the present investigation, whereas the minimum values in the decreasing order of 0.18 ± 0.02, 0.17 ± 0.04, 0.16 ± 0.04, and 0.07 ± 0.03 observed over XN, BJ, HH and KM, respectively during the winter season. This abrupt variation can be attributed to the fact that these are urban, industrial, and densely populated regions.
The fact that high convective activity makes the sand particles to lift into the atmosphere, anthropogenic aerosols produced by crop biomass burning (Lee et al., 2006; Tao et al., 2009; Li et al., 2010), and also the dust storm events initiates at the end of the spring season (transport coarse dust particles from north down to the east) which drastically changes the AOD distribution during the summer (Kaufman et al., 2005). Another reason for high values of AOD in summer is higher air temperatures which tend to hold abundant atmospheric water vapor facilitated the hygroscopic growth of aerosols (Masmoudi et al., 2003; Xin et al., 2005; Li et al., 2007). Similar results were also reported by several authors over China that increase in aerosol concentration which changes from season to season by regional anthropogenic and natural pollution (Pan et al., 2010; He et al., 2012; Luo et al., 2013).

**CORRELATION BETWEEN AOD AND CLOUD PARAMETERS**

Aerosols introduce a complicating factor due to their hygroscopic nature. Therefore, the aerosol-cloud interaction needs to be made by combining several methods (Alam et al., 2014; Kumar, 2014). Here we used AOD as a substitute for aerosol concentration and studied the relationships in detail with the help of one tailed distribution single paired student’s t-Test, slope which represents trend analysis and correlation studies for each set of parameters on monthly basis for different years throughout the study period (2003–2013). In this section, the relationship between MODIS derived AOD and cloud parameters such as WV, CF, COT, CER, CTP and CTT for the selected regions of China has been discussed and presented individually through statistical correlation analysis. The results with more details concerned to the relationship between AOD and various cloud properties are discussed in the following sections, except for WV, and CTP which are given in the SM (see S4, S5 and Fig. S1, Fig. S2, respectively).

**Correlation between AOD and CF**

The correlation plots showing the variation between AOD and CF with correlation coefficient either positive or negative over the selected regions of China for the period 2003–2013 is shown in Fig. 4. It is evident from Fig. 4 that the negative trend between AOD and CF occurs only when the AOD value have fallen below 0.4, which is in good agreement with the findings of Hoeve et al. (2012) and Alam et al. (2014). Also, CF increased with AOD from March to August and then decreases to reach its lowest value in February at all locations for the entire study period (figure not shown). In winter, relatively lower AOD values were observed due to reduction in the emissions from either natural or anthropogenic sources because of dry climatic conditions. Walcek (1994) and Myhre et al. (2007) have reported a good correlation between cloud cover, relative humidity (RH), and vertical velocity. The cloud cover exhibits a weak negative correlation with the potential temperature lapse rate, RH and vertical shear of the horizontal wind in the middle atmosphere (Walcek, 1994; Kaufman et al., 2005).

The statistical regression coefficients representing slope, correlation coefficient (r) and R² values obtained from the correlation between AOD and CF for the entire period are presented in Table 3. The calculated R² values were found to be maximum for KM (0.176) and minimum for BJ (0.0004). The correlation coefficients (r) were found to be negative for the regions BJ (-0.02), HH (-0.03), CD (-0.07), and LH (-0.08) and positive for the remaining regions where the maximum positive r value obtained for the region KM (0.42). The maximum positive slope of the trendline for the parameters AOD and CF was found for the region WH (0.352), whereas the least negative slope was noticed for the region CD (-0.366). The maximum probability obtained from the student t-Test between AOD and CF was found to be 0.855 for the region WH and minimum of 4.4E-72 for the region XN.

From Table 3, it is clear that the significant increase in the correlation between AOD and CF was found to be those regions which have more aerosol particles due to urban, industrial and domestic anthropogenic activities etc., with the influence of meteorological conditions. Albrecht (1989) and Kaufman and Fraser (1997) found that increase of aerosols may increase CF to a greater extent in regions highly loaded with WV, exhibiting low CCN than in environments laden with less WV. They indicated that this is due to the impact of a stronger updraft on the lifetime effect of clouds. Further, due to increase in aerosol concentrations, cloud cover increases and therefore, aerosol concentrations change the cloud properties. This is due to that the regions of low atmospheric pressure have more tendencies to create conditions necessary for cloud formation by accumulating aerosol particles and WV (Philipp et al., 2006; Sekiguchi et al., 2009; Wright et al., 2010).

**Correlation between AOD and COT**

Cloud optical thickness (COT) is a measure of attenuation of the light passing through the atmosphere due to the scattering and absorption by cloud droplets. It has a wide number of applications in radiative transfer, climate change, and hence in computing Earth’s radiation budget (Prasad et al., 2004). The statistical correlation between AOD and COT over the selected regions of China for the 10-year study period is shown in Fig. 5 and the corresponding regression coefficients obtained from the correlation were presented in Table 3. From Fig. 5 we show that the correlation was found to be negative where AOD is high, except for the urban, desert and mountainous regions. It is also noticed that a low COT was observed over the regions with high AOD. This might be due to its environmental and radiative factors (soot absorption and heating rates) that make some areas to host high aerosol loading and a low COT. During the study period, the COT was found to be increased with decrease in AOD at most of the regions except for the regions BJ, HH, XN and LH. The fact for the positive correlation between AOD and COT in the regions is due to their close proximity to the sea coast and is subjected to a component of sea and land breezes, as well as wet deposition, which contribute to a speedy clean-up of the atmosphere. Further, the regions close to the sea experiences...
Fig. 4. Correlation between AOD$_{550}$ and cloud fraction for the 12 selected sites in China during the period from 2003 to 2013. The statistical parameters obtained from the regression analysis are presented in Table 3.

A high amount of moisture as well, which ultimately results in an increased COT, as it depends highly on the moisture density and vertical depth of the cloud. Subsequently, a positive correlation was noticed between the two parameters (Alam et al., 2010).

The maximum positive R$^2$ value was noted for the region BJ (0.137) and the region UQ (0.0001) has the minimum R$^2$ value during the study period. The correlation coefficient (r) values were found to be negative in most of the regions and positive for the regions of BJ, HH, XN and LH. The positive r value was noticed to be maximum for the region BJ (0.37) and maximum negative for the region KM (−0.20) during the study period. The maximum slope for the trend was observed over BJ (0.013) which is significant at 95% confidence and the maximum (minimum) probability value obtained from the student’s t-Test was found to be 1.5E-43 (1.5E-97) for the region XN (CD) during the period of study. Quass et al. (2010) noticed a positive correlation between AOD and total cloud cover (TCC) using satellite retrievals and suggested that the dominant contribution to the AOD-TCC relationship can be attributed to aerosol swelling in regions where humidity is high and clouds are coincidentally found. It also provides us a direction that much of the AOD-TCC relationship seen in the satellite data was also carried by such a process, rather than the direct effects of the aerosols on the cloud fields themselves (Hoeve et al., 2012). The decreased COT was the presence of absorbing aerosols, which causes cloud droplets to evaporate making clouds too thin (decreased COT) results in negative correlation with AOD (Alam et al., 2014).
Table 3. Statistical parameters for the regression analysis with the monthly time series data obtained from AOD and cloud properties. The statistically significant trends at the 95% confidence level (p < 0.05) are presented in bold and italic and the rest are less significant. The values provided in the parenthesis for the slope (m) and coefficient of determination (R²) are intercept (c) and correlation coefficient (r), respectively for cloud parameter over different regions of China.

| Region | WV | CF | AOD versus | COT | CTP | CTT |
|--------|----|----|------------|-----|-----|-----|
|        | m (c) | R² (r) | \(|t|\) | m (c) | R² (r) | \(|t|\) | m (c) | R² (r) | \(|t|\) | m (c) | R² (r) | \(|t|\) |
| CC     | 0.017 (0.23) | 0.012 (0.11) | 7.0E-18 | 0.230 (0.09) | 0.053 (0.11) | 2.1E-46 | 0.013 (0.09) | 0.017 (0.11) | 3.3E-99 | -0.002 (0.28) | 0.002 (0.18) | 2.3E-55 | -0.0002 (0.36) | 0.012 (0.19) | 4.1E-87 | 0.001 (0.06) | 0.008 (0.00) |
| BJ     | 0.127 (0.20) | 0.436 (0.07) | 5.3E-19 | -0.176 (0.09) | 0.004 (0.18) | 1.6E-17 | 0.008 (0.09) | 0.005 (0.18) | 1.2E-99 | 0.013 (0.18) | 0.137 (0.18) | 2.5E-50 | -0.001 (1.19) | 0.462 (0.19) | 1.1E-92 | 0.002 (0.00) | 0.006 (0.00) |
| HH     | 0.056 (0.14) | 0.185 (0.08) | 4.3E-22 | -0.056 (0.23) | 0.009 (0.23) | 4.5E-42 | -0.003 (0.23) | 0.006 (0.23) | 1.9E-99 | 0.004 (0.14) | 0.053 (0.14) | 2.0E-50 | -0.0004 (0.46) | 0.260 (0.46) | 2.3E-89 | -0.004 (1.24) | 0.044 (0.21) |
| XN     | -0.005 (0.21) | 0.006 (0.11) | 1.7E-19 | 0.175 (0.11) | 0.063 (0.11) | 4.4E-72 | -0.001 (0.22) | 0.005 (0.22) | 3.1E-90 | 0.002 (0.20) | 0.008 (0.20) | 1.5E-43 | -0.0002 (0.31) | 0.068 (0.31) | 1.6E-99 | -0.008 (2.28) | 0.325 (0.57) |
| UQ     | 0.004 (0.30) | 0.001 (0.27) | 1.2E-27 | 0.079 (0.27) | 0.005 (0.27) | 9.7E-14 | 0.028 (0.30) | 0.123 (0.30) | 7.1E-96 | -0.002 (0.33) | 0.0001 (0.33) | 7.3E-50 | -0.0003 (0.46) | 0.102 (0.46) | 2.1E-73 | -0.008 (2.37) | 0.281 (0.53) |
| NJ     | 0.040 (0.62) | 0.096 (0.65) | 6.0E-18 | 0.084 (0.65) | 0.006 (0.65) | 7.977 | 0.064 (0.62) | 0.221 (0.62) | 1.6E-99 | -0.006 (0.84) | 0.02 (0.84) | 1.2E-64 | -0.001 (1.50) | 0.281 (1.50) | 3.7E-95 | -0.008 (2.83) | 0.048 (0.22) |
| WH     | 0.024 (0.48) | 0.029 (0.48) | 1.4E-21 | 0.354 (0.48) | 0.012 (0.48) | 0.855 | 0.040 (0.48) | 0.096 (0.48) | 1.9E-99 | -0.006 (0.86) | 0.012 (0.86) | 8.6E-72 | -0.0009 (1.29) | 0.160 (1.29) | 2.8E-96 | -0.007 (2.64) | 0.04 (0.02) |
| CD     | 0.005 (0.63) | 0.008 (0.63) | 2.0E-28 | -0.366 (0.63) | 0.005 (0.63) | 4.7E-13 | -0.0009 (0.63) | 0.008 (0.63) | 1.6E-97 | -0.008 (0.82) | 0.006 (0.82) | 1.5E-97 | 0.0001 (0.59) | 0.008 (0.59) | 8.7E-99 | -0.0008 (0.85) | 0.008 (0.09) |
| LH     | -0.031 (0.60) | 0.073 (0.50) | 2.0E-43 | -0.034 (0.50) | 0.006 (0.50) | 5.5E-6 | -0.029 (0.60) | 0.16 (0.60) | 2.9E-83 | 0.002 (0.88) | 0.008 (0.88) | 5.0E-68 | 0.0002 (0.36) | 0.023 (0.36) | 1.6E-64 | 0.002 (0.36) | 0.023 (0.15) |
| GZ     | 0.025 (0.56) | 0.017 (0.41) | 1.4E-49 | 0.319 (0.41) | 0.048 (0.41) | 5.3E-7 | -0.006 (0.56) | 0.005 (0.56) | 8.2E-89 | -0.002 (0.69) | 0.005 (0.69) | 2.9E-62 | -0.0002 (0.81) | 0.012 (0.81) | 6.4E-86 | -0.002 (1.08) | 0.003 (0.05) |
| NN     | -0.008 (0.56) | 0.005 (0.47) | 9.0E-48 | 0.085 (0.47) | 0.005 (0.47) | 1.3E-23 | -0.015 (0.56) | 0.023 (0.56) | 1.2E-85 | -0.003 (0.59) | 0.002 (0.59) | 1.0E-61 | 0.0003 (0.36) | 0.023 (0.36) | 1.6E-85 | 0.004 (0.43) | 0.026 (0.16) |
| KM     | 0.101 (0.01) | 0.372 (0.02) | 4.7E-45 | 0.295 (0.02) | 0.176 (0.02) | 2.2E-45 | 0.039 (0.01) | 0.230 (0.01) | 4.5E-96 | -0.008 (0.37) | 0.04 (0.37) | 1.3E-85 | -0.0008 (0.66) | 0.281 (0.66) | 4.1E-92 | -0.009 (2.54) | 0.203 (0.45) |

m—slope; c—intercept; R²—coefficient of determination; r—correlation coefficient; |t|—student’s t-Test probability.
Correlation between AOD and CER

The cloud effective radius (CER) is weighted mean values of size distribution of cloud droplets in the atmosphere. The correlation plots between AOD and CER shown in Fig. 6 found that the correlation is negative with maximum during the summer and minimum in the winter. The reduction in CER during the winter season could be due to the effect of aerosols on microphysical properties of cloud (Bhawar and Devara, 2010). Atmospheric circulation can decrease aerosol concentration and thereby decrease the probability of aerosol-cloud interaction. This in turn will reduce any impact that aerosol can have on cloud optical properties. In addition, aerosols originated from biomass burning inhibited the cloud droplet growth thereby caused an increase in the droplet residence time (Ackerman et al., 2000). This resulted in the low probability of warm rain since the cloud droplets attained higher altitude (Kumar, 2013).

The regression parameters obtained from the correlation between AOD and CER were tabulated in Table 3. The correlation coefficient (r) was found to be highly positive of 0.48 and 0.47 for the regions KM and NJ, respectively and high negative value of −0.40 for the region LH. The maximum slope of the trendline was noticed for the region NJ (0.064) which is 95% significant level and minimum of −0.0009 for CD where the trendline is almost a straight line parallel to abscissa (CER). The probability value was found to be small for BJ which is 1.2E-99 during the period of study. Thus it is clear in the present study that CER decreased with increased AOD. Kaufman et al. (2005) also reported a decreasing trend of CER with increasing AOD over the Atlantic Ocean. Recently, Tang et al. (2014) also found
similar results between AOD and CER over the open oceanic regions in the East part of China. However, the positive correlation was noticed over the land between AOD and CER which is due to the other processes such as microphysical and dynamical effects that are likely counteracting the indirect effect of aerosols on cloud droplets, which is defined as the well-known Twomey effect (Twomey, 1977).

Myhre et al. (2007) attributed the fact for the positive correlation between AOD and CER observed in the Mediterranean Sea. They hypothesized that as CTP decreases, AOD and CER increase, thus weakening the positive correlation between AOD and CER when CTP is large and vice-versa. Yuan et al. (2008) also revealed positive correlations between AOD and CER over the Gulf of Mexico and Eastern China, which they considered to be related to the effects of slightly soluble organic particles (SSO) and giant CCN. They explained that such particles can contribute to large AOD but fewer total cloud droplets and thus higher CER. Also, Gunaseelan et al. (2014) found that AOD is negatively correlated with CER for the four metro cities in India.

**Correlation between AOD and CTT**

In order to correlate atmospheric and surface properties properly, CTT data is needed to be analyzed accurately. Balakrishnaiah et al. (2012) observed that CTT plays a significant role in studies concerned with atmospheric radiation budget. The correlation plots between AOD at 550 nm and CTT during the period of study for the 12 regions were shown in Fig. 7. The CTT was found to be high in late autumn where as its low value occurred in summer season (figure not shown) for all the regions where AOD

![Cloud Effective Radius (µm)](image-url)

Fig. 6. Same as Fig. 4, but for cloud effective radius.
and CTT were negatively correlated. It is clearly depicted from Fig. 7 that CTT was observed to be increased with decrease in AOD (negative) at all regions except for the regions CC, BJ, LH and NN, where CTT increases with increase in AOD (positive). This positive correlation might be due to complex interlay among convection, boundary layer and large-scale cloud parameterization in those regions (Quass et al., 2010). The correlation coefficient (r) given in Table 3 between AOD and CTT showed strong negative correlation for the region XN (–0.57) followed by UQ (–0.53) and KM (–0.45). The maximum slope of the decreasing trendline for the period of 2003–2013 was found to be over region KM which is of –0.009 and very small negative slope of –0.0008 was obtained for the region CD where the trendline is almost straight line. The one tailed distribution single paired student’s t-Test between AOD and CTT was found to be negligible (almost zero) for all the regions of the study. Sekiguchi et al. (2009) and Xiong et al. (2009) reported that the aerosols acting on clouds change the cloud properties such as COT, CTT, etc. They also pointed out that the increasing aerosol number concentration due to anthropogenic and domestic activities may change the humidity profiles and thereby changing the CTT.

SUMMARY AND CONCLUSIONS

Cloud microphysics is substantially affected by aerosol loading and the resulting changes in the reflective properties of the clouds can significantly affect the regional and global radiation budget. The present investigation has revealed the spatial and temporal relationship between the AOD and different cloud parameters derived from MODIS.
data on board Terra satellite for the period from March 2003 to February 2013 over 12 selected metro cities in China. The major findings of the present study are:

1. AOD was found to be maximum (≥ 0.6) over North Chain Plain, East, West and Central parts of China, where as the minimum AOD of < 0.3 was observed over North and Southwest of China and the Tibet Plateau region. The AOD varies seasonally (monthly) from low in the winter months (January/February) to the high in the summer season (June/July) followed by spring and autumn seasons.

2. The high AOD over the selected regions in China may be attributed to the rapid urbanization results in increase in automobile transportations, industrial emissions, large number of construction activities, and also the increase in amount of aerosols produced from biomass burning. The dense population, geography affected by terrain and local sources, climate and economy are also closely related with the aerosol climatology over China.

3. In the present investigation, the correlation between AOD and CF was found to be comparatively lower at urban and industrial regions than at the coastal and desert areas where the correlation is noticed to be higher. We also found that the correlation between AOD and CF is negative only when the AOD fall below to 0.3. COT and AOD showed negative correlation for all the regions except in Beijing, Hohhot, Xining, and Guangzhou.

4. Positive correlation was observed between AOD and CER in urban and desert regions and negative over coastal locations of China. The positive correlation over urban and industrial areas is likely attributed to meteorological conditions and the general atmospheric circulations that favor the transport of pollutants and WV from southeast and eastern parts of China, leading to simultaneous increases in both AOD and CER.

5. The correlation between AOD and CTT was observed to be negative as CTT increased with decrease in AOD at all regions except Changchun, Beijing, Lhasa, and Nanning where CTT increases with increase in AOD. This is because of the fact that although there was a significant negative correlation between the AOD and CTT but the CTT was found to remain insensitive with respect to the changes in aerosol concentration.

ACKNOWLEDGEMENTS

This work was supported by the Strategic Priority Research Program (B) of the Chinese Academy of Sciences (Grant No. XDB05030104), the National Natural Science Foundation of China (Grant No. 41275128), the Natural Science Foundation of Jiangsu Province (Grant No. BK20140996), Qing Lan Project and the Priority Academic Program Development (PAPD) of Jiangsu Higher Education Institution. We gratefully acknowledge the MODIS science data support team for processing data via the GES-DISC interactive online visualization and analysis infrastructure (Giovanni), as a part of NASA. Thanks are also due to Mr. Liang Xuwei for the help in developing MATLAB codes to generate spatial maps for this study. The authors would like to thank Dr. James R. Campbell, the editor of the journal and the two anonymous reviewers for their insightful comments and constructive suggestions which in turn helped to improve the clarity and scientific content of the original manuscript.

SUPPLEMENTARY MATERIALS

Supplementary data associated with this article can be found in the online version at http://www.aqqr.org.

REFERENCES

Ackerman, A.S., Toon, O.B., Stevens, D.E., Heymsfield, A.J. and Ramanathan, V. (2000). Reduction of Tropical Cloudiness by Soot. Soot. Science 288: 1042–1047.

Alam, K., Iqbal, J., Blaschke, T., Qureshi, S. and Khan, G. (2010). Monitoring Spatio-temporal Variations in Aerosols and Aerosol Cloud Interaction over Pakistan Using MODIS Data. Adv. Space Res. 46: 1162–1176.

Alam, K., Khan, R., Blaschke, T. and Mukhtiar, A. (2014). Variability of Aerosol Optical Depth and Their Impact on Cloud Properties in Pakistan. J. Atmos. Sol. Terr. Phys. 107: 104–112.

Albrecht, B.A. (1989). Aerosols, Cloud Microphysics, and Fractional Cloudiness. Science 254: 1227–1230.

Anderson, T.L., Carlson, R.J., Shwartz, S.E., Knutti, R., Boucher, O., Rodhe, H. and Heintzenberg, J. (2003). Climate Forcing by Aerosols – A Hazy Picture. Science 300: 1103–1104.

Balakrishnaiah, G., Kumar, K.R., Reddy, B.S.K., Gopal, K.R., Reddy, R.R., Reddy, L.S.S. and Babu, S.S. (2012). Spatio-temporal Variations in Aerosol Optical and Cloud Parameters over Southern India Retrieved from MODIS Satellite Data. Atmos. Environ. 47: 435–445.

Bhawar, R.L. and Devara, P.C.S. (2010). Study of Successive Contrasting Monsoon (2001-2002) in Terms of Aerosol Variability over Tropical Station Pune, India. Atmos. Chem. Phys. 10: 29–37.

Chan, P.W. (2009). Comparison of Aerosol Optical Depth (AOD) Derived from Ground-based LIDAR and MODIS. Open Atmos. Sci. J. 3: 131–137.

Dong, Z.P., Yu, X., Li, X.M. and Dai, J. (2013). Analysis of Variation Trends and Causes of Aerosol Optical Depth in Shaanxi Province Using MODIS Data. Chinese Sci. Bull. (Atmos. Sci.), doi: 10.1007/s11434-013-5991-z.

Dutkiewicz, V.A., Alvi, S., Ghauri, B.N., Choudhary, M.I. and Husain, L. (2009). Black Carbon Aerosols in Urban Air in South Asia. Atmos. Environ. 43: 1737–1744.

Feingold, G., Eberhard, W.L., Veron, D.E. and Previdi, M. (2003). First Measurements of the Twomey Indirect Effect Using Ground-based Remote Sensors. Geophys. Res. Lett. 30: 1287, doi: 10.1029/2002GL016633.

Forest, C.E., Stone, P.H., Sokolov, A.P., Allen, M.R. and Webster, M.D. (2002). Quantifying Uncertainties in Climate System Properties with the Use of Recent Climate Observations. Science 295: 113–117.

Gunaseelan, I., Bhaskar, B.V. and Muthuchelian, K. (2014). The Effect of Aerosol Optical Depth on Rainfall with Reference to Meteorology over Metro Cities in India.
Environ. Sci. Pollut. Res. doi: 10.1007/s11356-014-2711-4.
Guo, X., Fu, D., Guo, X. and Zhang, C. (2014). A Case Study of Aerosol Impacts on Summer Convective Clouds and Precipitation over Northern China. Atmos. Res. 142: 142–157.
Hansen, J., Sato, M. and Ruedy, R. (1997). Radiative Forcing and Climate Response. J. Geophys. Res. 102: 6831–6864.
He, Q., Li, C., Geng, F., Lei, G. and Li, Y. (2012). Study on Long-term Aerosol Distribution over the Land of East China Using MODIS Data. Aerosol Air Qual. Res. 12: 304–319.
Hoeve, J.E.T., Remer, L.A. and Jacobson, M.Z. (2012). Microphysical and Radioactive Effect of Aerosols on Warm Clouds during the Amazon Biomass Burning Season as Observed by MODIS: Impact of Water Vapor and Land Cover. Atmos. Chem. Phys. 11: 3021–3036.
Intergovernmental Panel on Climate Change (IPCC) (2013) Climate Change 2013: The Physical Science Basis: Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change. Stokker, T.F., Qin, D., Plattner, G.K., Tignor, M., Allen, S.K., Boschung, J., Nauels, A., Xia, Y., Bex, V. and Midgley, P.M. (Eds.), Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA, 1535 pp.
Johnson, B.T., Shine, K.P. and Forster, P.M. (2004). The Semi-direct Aerosol Effect: Impact of Absorbing Aerosols on Marine Stratocumulus. Q. J. R. Meteorol. Soc. 130: 1407–1422.
Kaufman, Y.J. and Fraser, R.S. (1997). The Effect of Smoke Particles on Clouds and Climate Forcing. Science 277: 1636–1639.
Kaufman, Y.J., Koren, I., Remer, L.A., Rosenfeld, D. and Rudich, Y. (2005). The Effect of Smoke, Dust, and Pollution Aerosol on Shallow Cloud Development over the Atlantic Ocean. Proc. Nat. Acad. Sci. U.S.A. 102: 11207–11212.
Kim, B.G., Schwartz, S.E., Miller, M.A. and Min, Q.L. (2003). Effective Radius of Cloud Droplets by Ground-based Remote Sensing: Relationship to Aerosol. J. Geophys. Res. 108: 4740, doi: 10.1029/2003JD003721.
King, M.D., Kaufman, Y.J., Menzel, W.P. and Tanre, D. (1992). Remote Sensing of Cloud, Aerosol, and Water Vapor Properties from the Moderate Resolution Imaging Spectroradiometer (MODIS). IEEE Trans. Geosci. Remote Sens. 30: 2–27.
Knutti, R., Stocker, T.F., Joos, F. and Plattner, G.K. (2002). Constraints on Radiative Forcing and Future Climate Change from Observations and Climate Model Ensembles. Nature 416: 719–723.
Koren, I., Kaufman, Y.J., Remer, L.A. and Martins, J.V. (2004). Measurement of the Effect of Amazon Smoke on Inhibition of Cloud Formation. Science 303: 1342–1345.
Kumar, A. (2013). Variability of Aerosol Optical Depth and Cloud Parameters over North Eastern Regions of India Retrieved from MODIS Satellite Data. J. Atmos. Sol. Terr. Phys. 1004–101: 34–49.
Kumar, A. (2014). Long Term (2003-2012) Spatio-Temporal MODIS (Terra/Aqua Level 3) Derived Climatic Variations of Aerosol Optical Depth and Cloud Properties over a Semi Arid Urban Tropical Region of Northern India. Atmos. Environ. 83: 291–300.
Lee, K.H., Kim, Y.J. and Han, J.S. (2006). Characteristics of Aerosol Observed during Two Severe Haze Events over Korea in June and October 2004. Atmos. Environ. 40: 5146–5155.
Lee, S.S. and Penner, J.E. (2011). Dependence of Aerosol-cloud Interactions in Stratocumulus Clouds on Liquid-water Path. Atmos. Environ. 45: 6337–6346.
Levy, R.C., Remer, L.A. and Dubovik, O. (2007). Global Aerosol Optical Properties and Application to Moderate Resolution Imaging Spectroradiometer Aerosol Retrieval over Land. J. Geophys. Res. 112, D13210.
Li, B., Yuan, H., Niu, F. and Tao, S. (2010). Spatial and Temporal Variations of Aerosol Optical Depth in China during the Period from 2003 to 2006. Int. J. Remote Sens. 31: 1801–1817.
Li, C., Mao, J.T., Lau, A.K., Liu, X., Liu, G. and Zhu, I. (2003). Research on the Air Pollution in Beijing and Its Surroundings with MODIS AOD Products. China J. Atmos. Sci. 27: 869–880.
Li, Z. (2004). Aerosol and Climate: A Perspective from East Asia. Observation. In Theory and Modeling of Atmospheric Variability, Zhu, X. (Ed.), World Scientific Pub Co., p. 501–525.
Li, Z., Niu, F., Lee, K.H., Xin, J., Hao, W.M., Nordren, B., Wang, Y. and Wang, P. (2007). Validation and Understanding of Moderate Resolution Imaging Spectroradiometer Aerosol Products (C5) Using Ground-based Measurements from the Handheld Sun Photometer Network in China. J. Geophys. Res. 112, doi: 10.1029/2007JD008479.
Li, Z.Q., Niu, F., Fan, J., Liu, Y., Rosenfeld, D. and Ding, Y. (2011). Long-term Impacts of Aerosols on the Vertical Development of Clouds and Precipitation. Nat. Geosci. 12: 888–894.
Lohman, U. and Feichter, J. (2005). Global Indirect Aerosol Effects: A Review. Atmos. Chem. Phys. 5: 715–737.
Luo, Y., Zheng, X., Zhao, T. and Chen, J. (2013). A Climatology of Aerosol Optical Depth over China from Recent 10 years of MODIS Remote Sensing Data. Int. J. Climatol. 34: 863–870, doi: 10.1002/joc.3728.
Masmoudi, M., Chaabane, M., Tanre, D., Gouloup, P., Barel, L. and Elleuch, F. (2003). Spatial and Temporal Variability of Aerosol: Size Distribution and Optical Properties. Atmos. Res. 66: 1–19.
Myhre, G., Stordal, F., Johnsrud, M., Kaufman, Y.J., Rosenfeld, D., Storelmo, T. and Isaksen, I. (2007). Aerosol-cloud Interaction Inferred from MODIS Satellite Data and Global Aerosol Models. Atmos. Chem. Phys. 7: 3081–3101.
Pan, L., Che, H.Z., Geng, F.H., Xia, X.G., Wang, Y.Q., Zhu, C.Z., Chen, M., Gao, W. and Guo, J.P. (2010). Aerosol Optical Properties Based on Ground Measurements over the Chinese Yangtze Delta region. Atmos. Environ. 44: 2587–2596.
Philipp, F., Markus, F., Fritzsehe, L. and Petzold, A. (2006). Measurement of Ultrafine Aerosol Size Distributions by a Combination of Diffusion Screen Separators and Condensation Particle Counters. \textit{J. Aerosol Sci.} 37: 577–597.

Prasad, A.K., Singh, R.P. and Singh, A. (2004). Variability of Aerosol Optical Depth over Indian Subcontinent Using MODIS Data. \textit{J. Indian Soc. Remote Sens.} 32: 313–316.

Quass, J., Boucher, O., Bellouin, N. and Kinne, S. (2008). Satellite-based Estimate of the Direct and Indirect Aerosol Climate Forcing. \textit{J. Geophys. Res.} 113, doi: 10.1029/2007JD008962.

Quass, J., Stevens, B., Stier, P. and Lohmann, U. (2010). Interpreting the Cloud Cover Aerosol Optical Depth Relationship Found in Satellite Data Using a General Circulation Model. \textit{Atmos. Chem. Phys.} 10: 6129–6135.

Ranjan, R.R., Joshi, H.P. and Iyer, K.N. (2007). Spectral Variation of Total Column Aerosol Optical Depth over Rajkot: A Tropical Semi-arid Indian Station. \textit{Aerosol Air Qual. Res.} 7: 33–45.

Remer, L.A., Kaufman, Y.J., Tanre, D., Mattoo, S., Chu, D.A., Martins, J.V., Li, R.R., Ichoku, C., Levy, R.C., Kliedman, R.G., Eck, T.F., Vermote, E. and Holben, B.N. (2005). The MODIS Aerosol Algorithm, Products and Validation. \textit{J. Atmos. Sci.} 62: 947–973.

Rosenfeld, D. (2000). Suppression of Rain and Snow by Urban and Industrial Air Pollution. \textit{Science} 287: 1793–1796.

Satheesh, S.K. and Moorthy, K.K. (2005). Radiative Effects of Natural Aerosols: A Review. \textit{Atmos. Environ.} 39: 2089–2110.

Sekiguchi, M., Nakajima, T., Suzuki, K., Kawamoto, K., Higurashi, A., Rosenfeld, D., Sano, I. and Mukai, S. (2009). A Study of the Direct and Indirect Effects of Aerosols Using Global Satellite Data Sets of Aerosol and Cloud Parameters. \textit{J. Geophys. Res.} 108: 4699.

Tai, A.P.K., Mickley, L.J. and Jacob, D.J. (2010). Correlations between Fine Particulate Matter (PM2.5) and Meteorological Variables in the United States: Implications for the Sensitivity of PM2.5 to Climate Change. \textit{Atmos. Environ.} 44: 3976–3984.

Tang, J., Wang, P., Mickley, L.J., Xia, X., Liao, H., Yue, X., Sun, L. and Xia, J. (2014). Positive Relationship between Liquid Cloud Droplet Effective Radius and Aerosol Optical Depth over Eastern China from Satellite Data. \textit{Atmos. Environ.} 84: 244–253.

Tao, J., Wang, Z., Han, D., Li, S., Su, L. and Chen, L. (2009). Analysis of Crop Residue Burning and Tropospheric NO2 Vertical Column Density Retrieved from Satellite Remote Sensing in North China (in Chinese). \textit{China Environ. Sci.} 29: 1016–1020.

Twomey, S. (1977). Influence of Pollution on Shortwave Albedo of Clouds. \textit{J. Atmos. Sci.} 34: 1149–1152.

Walcek, C.J. (1994). Cloud Cover and Its Relationship to Relative Humidity during a Springtime Midlatitude Cyclone. \textit{Mon. Weather Rev.} 122: 1021–1035.

Wright, M.E., Dean, B., Atkinson, Zieman, L., Griffin, R., Hiranuma, N., Sarah, B., Lefer, B., Flynn, J., Rperna, Rappengluck, B., Luke, W. and Kelleye, P. (2010). Extensive Aerosol Optical Properties and Aerosol Mass Related Measurements during TRAMP/TexAQS 2006-Implications for PM Compliance and Planning. \textit{Atmos. Environ.} 44: 4035–4044.

Xin, J.Y., Wang, Y.S., Li, Z.Q., Wang, P.C., Hao, W.M., Nordgren, B.L. and Wang, S.G. Liu, G.G., Wang, L.L., Wen, T.X., Sun, Y. and Hu, B. (2005). Aerosol Optical depth (AOD) and Angstrom Exponent of Aerosols Observed by the Chinese Sun Hazemeter Network from August 2004 to September 2005. \textit{J. Geophys. Res.} 112: D05203, doi: 10.1029/2006JD007075.

Xin, L., Ern, T.W., Khoo, R., Yoon, A.K., Chew, B.N., Salina, S.V. and Liew, S.C. (2007). Characterization of Aerosol Optical Depth and Angstrom Exponent across Singapore from Sun Photometer Measurements, CRISP, Singapore.

Xiong, X., Chiang, K., Sun, J., Barnes, W.I., Guenther, B. and Salomonson, V.V. (2009). NASA EOS Terra and Aqua MODIS on-orbit Performance. \textit{Adv. Space Res.} 43: 413–422.

Yuan, T.L., Li, Z.Q., Zhang, R.Y. and Fan, J.W. (2008). Increase of Cloud Droplet Size with Aerosol Optical Depth: An Observation and Modeling Study. \textit{J. Geophys. Res.} 113, doi: 10.1029/2007JD008632.

Zhang, X.Y., Gong, S.L., Zhao, T.L., Arimoto, R., Wang, Y.Q. and Zhou, Z.J. (2003). Sources of Asian Dust and Role of Climate Change versus Desertification in Asian Dust Emission. \textit{Geophys. Res. Lett.} 30: 2272, doi: 10.1029/2003GL018206.

Received for review, August 26, 2014
Accepted, November 5, 2014
SUPPLEMENTARY MATERIAL

Correlation analysis between AOD and cloud parameters to study their relationship over China using MODIS data (2003-2013): Impact on cloud formation and climate change

Na Kang, Kanike Raghavendra Kumar*, Yan Yin, Yiwei Diao, Xingna Yu

Key Laboratory for Aerosol-Cloud-Precipitation of China Meteorological Administration, School of Atmospheric Physics, Nanjing University of Information Science and Technology, Nanjing 210044, Jiangsu, China.

Content

S1. Literature review
S2. Methodology
S3. Relationship between AOD and WV
S4. Relationship between AOD and CTP
Fig. S1. Relationship between AOD$_{550}$ and water vapor for the 12 selected regions in China during the period 2003–2013.
Fig. S2. Same as Fig. S1, but for cloud top pressure.

*Corresponding author: Tel.: +86-25-58731592, Fax: +86-25-5869971, e-mail: kanike.kumar@gmail.com; kkrkumar@nuist.edu.cn
S1. Literature review

Alam et al. (2010) analyzed the effect of aerosols on clouds and showed enhanced formation of clouds was due to both smoke and pollution over Karachi and Lahore in Pakistan. Balakrishnaiah et al. (2012) noticed high AOD during the summer-monsoon seasons over major cities in southern India and studied the relationship to various cloud parameters. Yi et al. (2012) found an increasing variation in cloud fraction (CF) with the increase of AOD over ocean regions under observation, while the reverse result was noticed in the model simulation. Hoeve et al. (2012) investigated an increase in COT with increasing AOD at low AODs, and a decrease in COT with increasing AOD at higher AODs. This increase was attributed to a combination of microphysical and dynamical effects, whereas, the decrease was due to the dominance of radiative effects that thin and darken clouds.

Kumar (2013) also analyzed that AOD and CF correlation increases for those regions which have more particulate pollution due to dust, biomass, industrial and domestic activities over the northeast areas of India. Another study by Kumar (2014) found that AOD showed positive correlation with CF and COD and negative correlation with cloud top temperature (CTT) and cloud top pressure (CTP) over an urban region of northern India (Delhi) for the period 2003–2012. Alam et al. (2014) have recently studied the relationship between AOD and cloud properties in different locations of Pakistan. They noticed an increasing trend in CF with AOD over urban regions during the period of study from 2001 to 2011 as well as investigated the impact of aerosols on warm and cold clouds. Also very recently, Tang et al. (2014) showed positive correlation between AOD and cloud effective radius (CER) over open oceanic regions of East China, suggesting that the influence of background weather conditions and general circulations need to be considered when studying the interactions between aerosol and cloud.

S2. Methodology

The Ångström exponent (AE) is a qualitative indicator of aerosol particle size (Kaufman and Nakajima, 1993). The value of AE less than 1 indicates particle size dominated by coarse-mode aerosols with radii larger than 0.5 \( \mu \text{m} \), which are usually associated with dust and sea salt, and values larger than 2 indicates particle size dominated by fine-mode aerosols with radii less than 0.5 \( \mu \text{m} \) that are usually associated with urban pollution and biomass burning (Eck et al., 1999). AE from the MODIS product can be employed for qualitative judgment of the aerosol mode (Ångström, 1961). Assuming the size distribution of aerosol particles fits Junge-distribution, the relationship between AOD and AE can be expressed as:

\[ \tau_a(\lambda) = \beta \lambda^{-\alpha} \]

where \( \tau_a(\lambda) \) is the AOD at \( \lambda \), \( \beta \) is Ångström turbidity and \( \alpha \) is Ångström wavelength exponent inversely related to the effective radius of aerosol particles. MODIS AOD at 550 nm can be obtained by interpolation between AODs at 470 nm and 660 nm based on the above formula.
S3. Relationship between AOD and water vapor (WV)

The behavior of aerosols in response to changes in WV was investigated and it provided an opportunity to understand the impact of aerosols on Earth’s atmosphere (Myhre et al., 2007). The time series plot for AOD and WV is shown in Fig. S1 for all the selected regions in China during the period 2003–2013 represents increasing and decreasing pattern simultaneously. This is in accordance with the findings of Ranjan et al. (2007), Balakrishnaiah et al. (2012), and Kumar et al. (2009) reported over Indian subcontinent and Alam et al. (2010, 2014) for Pakistan. The direct effect results in radiation scattering due to an increase in aerosol particle-size, accompanied by the uptake of WV. Changes in water uptake of aerosols can therefore, lead to changes in both direct and indirect radiative forcing on climate (IPCC, 2007). El-Askary and Kafatos (2008) have found that aerosols cause a reduction in cloud droplet size and hence lead to suppression in precipitation.

The statistical regression data obtained from the correlation analysis between AOD and WV were shown in Table 3. It clearly demonstrates from Table 3 that the calculated coefficient of determination ($R^2$) values clearly shows the positive correlations for the above relation. The highest positive $R^2$ value was found in the increasing order for the regions BJ (0.436), KM (0.372) and HH (0.185) with its lowest value in the decreasing order for the regions CD (0.008), XN (0.006) and NN (0.005). Similarly, the correlation coefficient ($r$) values were noticed to be positive for all the regions, except for the regions XN, LH and NN where $r$ value found to be negative. The highest positive correlation coefficient value was noticed to be 0.66 and 0.61 for the regions BJ and KM, respectively followed by HH (+0.43) and NJ (+0.31) and maximum negative $r$ value over region LH (-0.27). The maximum slope of the trendline for the all the regions computed was found to be 0.127 and 0.101 between AOD and WV observed for the regions BJ and KM, respectively were statistically significant at 95% confidence level ($p<0.05$). The student’s t-Test probability was performed to the data and the maximum (minimum) probability $|t|$ was found to be 6.0E-18 (1.4E-49) for NJ (GZ). This uncertainty may occur in the AOD due to contamination by both low and high altitude cirrus clouds. Lee et al. (2009) concluded that cirrus clouds can increase the uncertainty in measured AOD by up to 20%.

The water absorbing ability of aerosols (hygroscopic nature) depends upon the particular mixing of different types of aerosols particles as well as on the meteorological conditions (Kaufman et al., 2005; Kaufman and Koren, 2006; Aloysius et al., 2009). Lee et al. (2009) suggested that water uptake of atmospheric aerosols can alter both the size and composition of particles and hence their optical particles. Wright et al. (2010) and Xie et al. (2011) depicted that the radiative forcing on climate significantly influenced by the changes in water uptake behavior of aerosols and hence cloud formation. Guo et al. (2014) and Luo et al. (2013) concluded that natural and anthropogenic aerosols over China play an important role in influencing the convective cloud formation and hence causing climatic implications to the overall hydrological cycle.
Fig. S1. Relationship between AOD$_{550}$ and water vapor for the 12 selected sites in China during the period of 2003–2013.
S4. Relationship between AOD and CTP

The time series variation between AOD and CTP shown in Fig. S2 also represents the similar variation with increase in AOD there was a significant decrease in the CTP value over the selected regions of China. It is clear from Fig. S2 that CTP reaches a maximum value during January with a minimum value in July, where the AOD pattern is exactly opposite to that of CTP for most of the regions (see Table1). The correlation coefficient between AOD and CTP for various regions in China over the period of study is shown in Table 3. The $R^2$ value noticed to be maximum for region of BJ (0.462) and minimum over CD (0.008). Out of 12 selected regions in China, a strong negative correlation coefficient was found over 9 regions of which the maximum negative r value obtained for the region B (-0.68) followed by the regions NJ (-0.53), KM (-0.53), and HH (-0.51). Earlier researchers have reported that except for some regions of low AOD, CTP decreased in most of the areas (higher cloud altitude) as AOD increased (Alam et al., 2014 and references therein). This might have resulted from the suppression of the precipitation by increasing cloud lifetime and thus also affecting the cloud albedo and changing the CTP (Ali et al., 2013). Further, Ramanathan et al. (2001) and Lee et al. (2009) suggested since the CER increases with decrease of CTP and thereby, decreasing the CTP with increasing AOD.

A very low slope of the decreasing trend was found in most of the regions indicates the trend is almost a straight line and the maximum (minimum) slope of the decreasing trendline was -0.001 (-0.0008) noticed for the regions BJ and NJ (XN) which is 95% significant level. The probability value was found to be low (high) for XN (LH) which is 1.6E-99 (1.6E-64) during the period of study. Alam et al. (2010) investigated that at lower latitudes, there was a significant decrease in CTP in relation to AOD (i.e., negative correlation), and while at mid latitudes this decrease was only moderate. Our analysis indicated a strong negative correlation for AOD with CTP in the southern regions and a moderate positive correlation in the northern regions of China which is in agreement with the previous findings reported for south Asia. This co-variation of AOD with CTP may be attributed to large-scale meteorological variations. Koren et al. (2005) and Tripathi et al. (2007) reported the relation between vertical wind velocity and CTP. They suggested that the relationship is less influenced by meteorology during winter due to reduced updraft, whereas, during summer-monsoon season the aerosol effect is facilitated by favorable meteorological condition, due to strong updraft.
Fig. S2. Same as Fig. S1, but for cloud top pressure.
REFERENCES

Alam, K., Iqbal, J., Blaschke, T., Qureshi, S. and Khan, G. (2010). Monitoring spatio-temporal variations in aerosols and aerosol cloud interaction over Pakistan using MODIS data. *Adv. Space Res.* 46: 1162–1176.

Alam, K., Khan, R., Blaschke, T. and Mukhtiar, A. (2014). Variability of aerosol optical depth and their impact on cloud properties in Pakistan. *J. Atmos. Sol. Terrs. Phys.* 107: 104–112.

Alloysius, M., Mohan, M., Babu, S.S., Parameswaran, K. and Moorthy, K.K. (2009). Validation of MODIS derived aerosol optical depth and an investigation on aerosol transport over the South East Arabian Sea during ARMEX-II. *Anna. Geophys.* 27: 2285–2296.

Ångström, A. (1961). Techniques of determining the turbidity of the atmosphere. *Tellus* 13: 214–223.

Balakrishnaiah, G., Kumar, K.R., Reddy, B.S.K., Gopal, K.R., Reddy, R.R., Reddy, L.S.S. and Babu, S.S. (2012). Spatio-temporal variations in aerosol optical and cloud parameters over southern India retrieved from MODIS satellite data. *Atmos. Environ.* 47: 435–445.

Eck, T.F., Holben, B.N., Reid, J.S., Dubovik, O., Smirnov, A. and O’Neill, N.T., et al. (1999). Wavelength dependence of the optical depth of biomass burning urban and desert dust aerosols. *J. Geophys. Res.* 104: 31333–31349.

El-Askary, H. and Kafatos, M. (2008). Dust storm and black cloud influence on aerosol optical properties over Cairo and the great delta region Egypt. *Int. J. Remt. Sens.* 29: 7199–7211.

Guo, X., Fu, D., Guo, X. and Zhang, C. (2014). A case study of aerosol impacts on summer convective clouds and precipitation over northern China. *Atmos. Res.* 142: 142–157.

Hoeve, J.E.T., Jacobson, M.Z. and Remer, L.A. (2012). Comparing results from a physical model with satellite and in situ observations to determine whether biomass burning aerosols over the Amazon brighten or burn of clouds. *J. Geophys. Res.* 117: 19.

Intergovernmental Panel on Climate Change (IPCC) (2007) Climate change 2007: The physical Science Basis: Contribution of Working Group I to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change. Solomon, S., Qin, D., Manning, M., Chen, Z., Marquis, M., Averty, K.B., Tignor, M. and Miller, H.L. Cambridge University Press, Cambridge/New York, pp.996.

Kaufman, Y.J. and Koren, I. (2006). Smoke and pollution aerosol effect on cloud cover. *Science* 313:655–658.

Kaufman, Y.J., Koren, I., Remer, L.A., Rosenfeld, D. and Rudich, Y. (2005). The effect of smoke, dust, and pollution aerosol on shallow cloud development over the Atlantic Ocean. *Proc. Natl. Acad. Sci.* 102: 11207–11212.

Kaufman, Y.J. and Nakajima, T. (1993). Effect of Amazon smoke on cloud microphysics and albedo-analysis from satellite imagery. *J. Appl. Meteorol.* 32: 729–744.
Koren, I., Kaufman, Y.J., Rosenfeld, D., Remer, L.A. and Rudich, Y. (2005). Aerosol invigoration and restructuring of Atlantic convective clouds. Geophys. Res. Lett. 32: L14828, http://dx.doi.org/10.1029/2005GL023187.

Kumar, A. (2013). Variability of aerosol optical depth and cloud parameters over North Eastern regions of India retrieved from MODIS satellite data. J. Atmos. Sol. Terrs. Phys. 100-101: 34–49.

Kumar, K.R., Narasimhulu, K., Reddy, R.R., Gopal, K.R., Reddy, L.S.S., Balakrishnaiah, G., Moorthy, K.K. and Babu, S.S. (2009). Temporal and spectral characteristics of aerosol optical depths in a semi-arid region of Southern India. Sci. Tot. Environ. 407: 2673–2688.

Lee, S., Ghim, Y.S., Kim, S. and Yoon, S. (2009). Seasonal characteristics of chemically apportioned aerosol optical properties at Seoul and Gosan, Korea. Atmos. Environ. 43: 1320–1328.

Luo, Y., Zheng, X., Zhao, T. and Chen, J. (2013). A climatology of aerosol optical depth over China from recent 10 years of MODIS remote sensing data. Int. J. Climatl. doi:10.1002/joc.3728.

Myhre, G., Stordal, F., Johnsrud, M., Kaufman, Y.J., Rosenfeld, D., Storelvmo, T. and Isaksen, I. (2007). Aerosol-cloud interaction inferred from MODIS satellite data and global aerosol models. Atmos. Chem. Phys. 7: 3081–3101.

Ramanathan, V., Crutzen, P.J., Kiehl, J.T. and Rosenfeld, D. (2001). Aerosols, climate, and the hydrological cycle. J. Geophys. Res. 294: 21119–21124.

Ranjan, R.R., Joshi, H.P. and Iyer, K.N. (2007). Spectral variation of total column aerosol optical depth over Rajkot: a tropical semi-arid Indian station. Aero. Air Qual. Res. 7: 33–45.

Tang, J., Wang, P., Mickley, L.J., Xia, X., Liao, H., Yue, X., Sun, L. and Xia, J. (2014). Positive relationship between liquid cloud droplet effective radius and aerosol optical depth over Eastern China from satellite data. Atmos. Environ. 84: 244–253.

Tripathi, S.N., Pattnaik, A. and Dey, S. (2007). Aerosol indirect effect over Indo-Gangetic Plain. Atmos. Environ. 41: 7037–7047.

Wright, M.E., Dean, B., Atkinson, Ziembia, L., Griffin, R., Hiranuma, N., Sarah, B., Lefer, B., Flynn, J., Rperna, Rappengluck, B. and Luke, W., et al. (2010). Extensive aerosol optical properties and aerosol mass related measurements during TRAMP/TexAQS 2006- Implications for PM compliance and planning. Atmos. Environ. 44: 4035–4044.

Xie, Young, Yan, Zhang, Xiong X, Qu John, K. and Che, H. (2011). Validation of MODIS aerosol optical depth product over China using CARSNET measurements. Atmos. Environ. 45:5970–5978.

Yi, B., Yang, P., Bowman, K.P. and Liu, X. (2012). Aerosol-cloud-precipitation relationships from satellite observations and global climate model simulations. J. Appl. Remt. Sens. 6: 063503, http://dx.doi.org/10.1117/1.JRS.6.063503.