Aerosol daytime variations over North and South America derived from multiyear AERONET measurements

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[1] This study analyzes the daytime variation of aerosol with seasonal distinction by using multiyear measurements from 54 of the Aerosol Robotic Network (AERONET) sites over North America, South America, and islands in surrounding oceans. The analysis shows a wide range of daytime variability of aerosol optical depth (AOD) and Ångström exponent depending on location and season. Possible reasons for daytime variations are given. The largest AOD daytime variation range at 440 nm, up to 75%, occurs in Mexico City, with maximum AOD in the afternoon. Large AOD daytime variations are also observed in the polluted mid-Atlantic United States and West Coast with maximum AOD occurring in the afternoon in the mid-Atlantic United States, but in the morning in the West Coast. In South American sites during the biomass burning season (August to October), maximum AOD generally occurs in the afternoon. But the daytime variation becomes smaller when sites are influenced more by long-range transported smoke than by local burning. Islands show minimum AOD in the morning and maximum AOD in the afternoon. The diverse patterns of aerosol daytime variation suggest that geostationary satellite measurements would be invaluable for characterizing aerosol temporal variations on regional and continental scales. In particular, simultaneous measurements of aerosols and aerosol precursors from a geostationary satellite would greatly aid in understanding the evolution of aerosol as determined by emissions, chemical transformations, and transport processes.

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1. Introduction

[2] Tropospheric aerosols have large spatial and temporal variations that are controlled by changing emissions from diverse origins, by meteorological processes on various scales, by chemical evolution, and by removal processes. The characteristic time scale of variation of aerosol optical

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depth (AOD) is about 3 h in remote regions, but can be less than 1 h near the emission sources [Anderson et al., 2003]. High spatial and temporal resolution measurements of aerosol loading and emissions are essential for improving particulate matter (PM) air quality forecasts [Kondragunta et al., 2008; Yang et al., 2011]. Aerosol daytime variations, in combination with changing geometry of Sun and surface reflectance, could lead to large daytime variations of aerosol radiative forcing [Yu et al., 2004]. Such variations of aerosol forcing need to be adequately represented in a model in order to realistically assess atmospheric responses to the radiative forcing, such as the atmosphere-surface interactions and the evolution of the atmospheric boundary layer [Yu et al., 2002]. Aerosols interact with clouds on the cloud lifetime scales which are significantly less than an hour. Finally, aerosol variations need to be taken into account when comparing different observations or integrating observations and models [Anderson et al., 2003, 2005]. For all these reasons, high temporal resolution aerosol measurements are needed and the daytime variations of aerosol loading need to be quantified.

[3] Surface networks and aircraft missions have made progress toward quantifying aerosol daytime variations [e.g.,

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Kaufman et al., 2000; Smirnov et al., 2002; Delene and Ogren, 2002; Anderson et al., 2003; Pandithurai et al., 2007; Michalsky et al., 2010; Redemann et al., 2005, Wang et al., 2004]. However, such studies are limited in spatial extent and/or longevity. Although polar orbiting satellites can survey the entire globe with high spatial resolution, they can only sample a particular location once a day. The daytime variations of aerosols on a large spatial scale can however be measured from geostationary earth orbit [e.g., Wang et al., 2003; Prados et al., 2007; Zhang and Kondragunta, 2008]. The major advantage of a geostationary measurement is its regional and continental coverage with high time and space resolution, which surface and aircraft measurements can never achieve. Current geostationary satellite sensors, such as The Geostationary Operational Environmental Satellites (GOES), were not designed to retrieve aerosol information, in part because of their coarse spatial resolution and limited wavelength band. As the result, the aerosol products from GOES have never reached the same level of accuracy and quality as the aerosol products from the EOS era polar orbiting missions. The U.S. National Research Council [National Research Council, 2007] has recommended the Geostationary Coastal and Air Pollution Events (GEO-CAPE) mission for the coming decade to advance science and meet societal needs in relation to atmospheric pollution chemistry, climate forcing, and coastal ecosystems. This mission offers an opportunity to design a geostationary satellite measurement of daytime variations aerosols and precursor gases with improved accuracy to advance the understanding of aerosol processes and aerosol effects on climate and air quality.

[4] As part of a NASA led effort to define the science requirements for the aerosol component of the GEO-CAPE mission [Fishman et al., 2012], this study analyzes the daytime variation of aerosol with seasonal distinction by using multiyear measurements from 54 of the Aerosol Robotic Network (AERONET) sites over North America, South America, and islands of the surrounding oceans (i.e., within the planned geographical coverage of GEO-CAPE). Both aerosol loading and size/type, as characterized by AOD and Ångström exponent (AE), respectively, are examined. The rest of paper is organized as follows. Section 2 gives an overview of major factors contributing to aerosol daytime variations to facilitate later discussions. Section 3 describes the AERONET data sets and method of calculating daytime variations. Section 4 presents the spatial patterns of aerosol daytime variation in the study domain and then discusses in more detail the aerosol daytime variation in several representative regions or sites. Major conclusions and implications for the GEO-CAPE mission are summarized in section 5.

2. Factors Contributing to Aerosol Daytime Variations

[5] Daytime variation of AOD and AE can be attributed to such factors as emissions, meteorological conditions, photochemical activities, and relative humidity (RH), among others. In what follows, we briefly describe several of these major factors. In reality, several factors usually work together to determine the aerosol daytime variation.

2.1. Emission

[6] Daytime variations of particle emissions directly control variations of AOD and probably AE, particularly in source regions. For example, biomass burning in South America is generally more active in the afternoon than in the morning [Prins et al., 1998; Giglio, 2007; Vermote et al., 2009] suggesting that AOD in smoke source regions is also higher in the afternoon. Over urban areas, aerosol and its precursor emissions are larger during rush hours than non–rush hours, contributing to the AOD and AE diurnal variation.

2.2. Meteorology

[7] Meteorological conditions, in particular those associated with mesoscale circulations, control the transport, evolution, and removal of aerosols on a daily time scale. For example, the land-sea breeze and mountain-valley circulations resulting from differential heating between land and sea and between mountain and valley, respectively, can play an important role in diluting or accumulating aerosols. The daytime sea breeze would bring maritime air into the continental boundary layer, which may lower the aerosol loading and increase the size of aerosol. The nighttime land breeze would bring continental air from inland to coastal area, possibly resulting in accumulation of aerosols in the coastal area. Similarly, the daytime upslope flow would bring polluted air from foothill to relatively pristine hilltops, and result in an increase of AOD over the day and a peak in the afternoon on the hilltop. Rain out and wash out are major scavenging mechanisms for aerosols. Therefore, the diurnal variation of clouds and precipitation would regulate diurnal variation of aerosols.

2.3. Photochemistry

[8] Secondary aerosol, such as sulfate, nitrate, and some organic aerosols are produced from precursor gases through photochemical processes. Such photochemical production rates for aerosols are determined by diurnal varying photodissociation frequencies that increase with increasing solar radiation and sometimes temperature. Aqueous chemistry is also one of the major chemical pathways for the formation of aerosols such as sulfate.

2.4. Hygroscopic Growth

[9] Hydrophilic aerosols, like sulfate, sea salt, nitrate, and some types of carbonaceous aerosol, can grow when the ambient relative humidity (RH) increases [Seinfeld and Pandis, 1998; Bian et al., 2009]. An increase of RH increases the particle size and hence the cross section of particle interacting with solar radiation, leading to an increase of AOD and decrease of AE. This process is highly nonlinear, with the rate of particle growth much higher at high RH than at low RH. Dust, black carbon, and some organic carbon aerosols are, by contrast, largely hydrophobic and their size change little with variation of RH.

3. AERONET Data and Analysis Method

[10] AERONET is a federated international ground-based global network established for characterizing aerosol optical properties and validating aerosol satellite retrievals [Holben et al., 1998]. The network started in 1993 and has since been expanded to more than 500 sites globally over nearly

Table 1. Latitude, Longitude, and Elevation Information of 54 AERONET Stations

Site Name	Longitude (Decimal Degrees)	Latitude (Decimal Degrees)	Elevation (m)
Abracos Hill	-62.4	-10.8	200
Alta Floresta	-56.1	-9.9	277
Arica	-70.3	-18.5	25
Bondville	-88.4	40.1	212
BSRN BAO Boulder	-105.0	40.0	1604
Belterra	-55.0	-2.6	70
Bermuda	-64.7	32.4	10
Bonanza Creek	-148.3	64.7	150
Bratts Lake	-104.7	50.3	586.7
Cartel	-71.9	45.4	300
CCNY	-73.9	40.8	100
Ceilap-BA	-58.5	-34.6	10
COVE	−75.7 • • • • • • • • • • • • • • • • • • •	36.9	37
Cuiaba-Miranda	-56.0	-15.7	210
Campo Grande	-54.6	-20.45	500
Cart Site	-97.5	36.6	318
Coconut Island	-157.8	21.4	0
Cordoba-CETT	-64.5	-31.5	730
Dry Tortugas	-82.9	24.6	0 264
Egbert Fresno	-79.8 -119.8	44.2 36.8	0
GSFC	-119.8 -76.8	39.0	87
HJAndrews	-76.8 -122.2	39.0 44.2	830
Halifax	-63.6	44.6	65
Howland	-68.7	45.2	100
KONZA EDC	-96.6	39.1	341
Kelowna	-119.4	50.0	344
La Jolla	-117.3	32.9	115
La Parguera	-67.0	18.0	12.4
Lanai	-156.9	20.7	20
MD Science Center	-76.6	39.3	15
Maricopa	-112.0	33.1	360
Mauna Loa	-155.6	19.5	3397
Mexico City	-99.2	19.3	2268
Midway Island	-177.4	28.2	20
Missoula	-114.1	46.9	1028
Monterey	-121.9	36.6	50
Railroad Valley	-116.0	38.5	1435
Rimrock	-117.0	46.5	824
Rio Branco	-67.9	-10.0	212
Rogers Dry Lake	-117.9	34.9	680
SERC	-76.5	38.9	10
San Nicolas	-119.5	33.3	133
Sao Paulo	-46.7	-23.6	865
Saturn Island	-123.1	48.8	200
Sevilleta	-106.9	34.4	1477
Sioux Falls	-96.6	43.7	500
Table Mountain CA	-117.7	34.4	2200
Tahiti	-149.6	-17.6	98
Trelew	-65.3	-43.2	15
Tucson	-111.0	32.2	779
Walker Branch	-84.3	36.0	365
Wallops	-75.5	37.9 53.0	10
Waskesiu	-106.1	53.9	550

two decades. Typically most Cimel Sun-sky radiometers deployed by AERONET measure the direct solar irradiances in wavelength channels 340, 380, 440, 500, 675, 870, 940, and 1020 nm (some have an additional channel at 1640 nm) with a nominal sampling frequency of 15 min (higher frequency in early morning and late afternoon in order to attempt Langley calibrations). Among direct sun channels, the 940 nm one is designed to estimate total precipitable water content and the remaining seven are used to retrieve AOD. An automatized and computerized cloud-screening

algorithm [Smirnov et al., 2000] is applied after AOD is calculated. The typical uncertainty in AOD for Level 2 AERONET data is ± 0.01 to ± 0.02 , with the larger errors appearing in ultraviolet bands [Eck et al., 1999]. The wavelength (λ) dependence of AOD, is characterized by Ångström exponent (AE) with the classical equation AOD(λ) $\sim \lambda^{-AE}$ [Ångström, 1929]. AE can be used as a proxy for aerosol size, with a value greater than 1 indicating fine mode (submicron radius) pollution and biomass burning aerosols and a value less than \sim 0.6 indicating coarse mode (supermicron radius) dust and sea-salt aerosols. In this study, we use only Version 2 Level 2 AERONET data, and derive AE from a linear fitting of lnAOD versus $\ln \lambda$, using measurements at the 4 CIMEL wavelengths in the range of 440–870 nm, following Eck et al. [1999].

[11] A total of 54 AERONET sites, mainly located in both South America and North America, and on islands in the surrounding oceans, were selected for this study. Table 1 lists latitude, longitude, and elevation for these sites. All these sites have at least two years of measurements available after 1997 (note that interference filter type was changed in 1997, with significant improvement in filter transmittance stability). Measurements prior to 1997 are excluded to retain only measurements with the highestquality calibration. We consider only the data that fall within the ranges of 0.01 < AOD < 5 and 0 < AE < 3 to eliminate unrealistic measurements. 92% of the data fall within these ranges for all sites. Given that the sample frequency and total number of measurements differ from site to site, all individual observations in a day are expressed as the departure (percentage) from the daily mean to avoid sampling number issues [Smirnov et al., 2002]. The calculation of diurnal average departure (percentage) for each season of AOD and AE for each AERONET site is as follows: (1) compute hourly mean AOD and AE by averaging all available instantaneous measurements within 1 h, for example, between 10:30 and 11:30 local time for each day; (2) calculate the daily mean by averaging all available hourly means, excluding days with less than five hourly means: (3) calculate percentage departures of individual hourly observations from the daily mean; (4) derive seasonal mean of hourly departure (percentage) by aggregating all hourly departures from the daily mean within an hour in a given season. We divide the data into the usual four seasons, namely December-February (DJF), March-May (MAM), June-August (JJA), and September-November (SON), except as otherwise specified. Daytime variation range (referred to as DVR, in percentage) is defined as the difference between the maximum and minimum hourly departure (in percentage) in a season. DVR combined with seasonal mean AOD and AE can be used to approximately estimate the absolute range of change over a day.

4. Results

4.1. Spatial Patterns of Aerosol Daytime Variations

[12] Figures 1 and 2 give an overview of daytime variations of AOD and AE, respectively, in all 54 AERONET sites on a seasonal basis. In Figures 1 and 2, seasonal means of AOD and AE are represented by different colors; DVRs by the size of triangle, and the occurring time of peak AOD

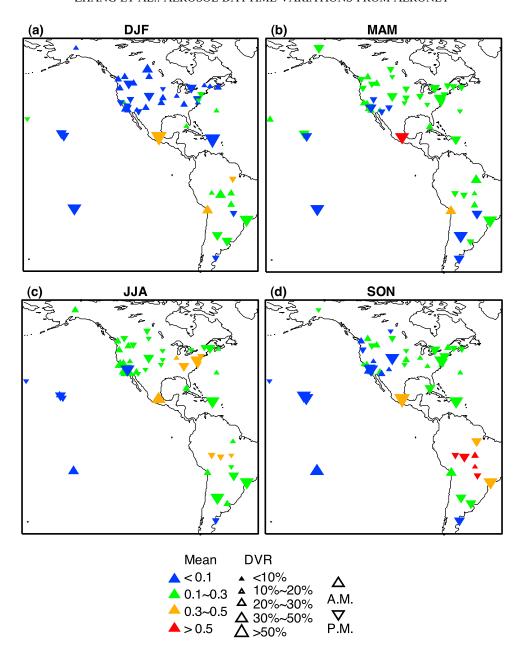


Figure 1. Geographical distributions of seasonal mean aerosol optical depth (AOD) at 440 nm (aggregated into 4 bins and marked with different colors), AOD daytime variation range (DVR is defined as a difference of maximum and minimum hourly percentage departure from the daily mean AOD, with its value being represented by size of triangle), and occurring time of maximum AOD (with upside triangle for morning and downside triangle for afternoon).

or AE by the direction of triangle (see Figures 1 and 2 legends for details). Figures 1 and 2 show a wide range of aerosol daytime variations, in terms of both DVR and occurring time of maximum value, depending on location and season. Mexico City has the largest AOD and the highest AOD daytime variations throughout the year, with DVR of 30–50% or higher. The maximum AOD occurs in the morning in summer and in the afternoon in other seasons.

[13] In the eastern part of the United States where industrial pollution dominates, AOD is generally highest in summer and lowest in winter. The high summertime AOD is

associated with high relative humidity, active photochemistry, and stagnant atmospheric circulations [Husar et al., 1981; Bian et al., 2010]. The DVR for AOD is generally larger than 10%, with the highest value about 30%. Maximum AOD for each season occurs in the afternoon. The daytime variations for AE in the eastern United States are generally small (less than 10%), particularly in summer (\sim 5%).

[14] In the western part of the United States, AOD DVRs are generally comparable to those in the eastern part of the United States in summer but smaller in other seasons. Given that the mean AOD is smaller in the West than in the East, the absolute daytime variation of AOD in the West is smaller

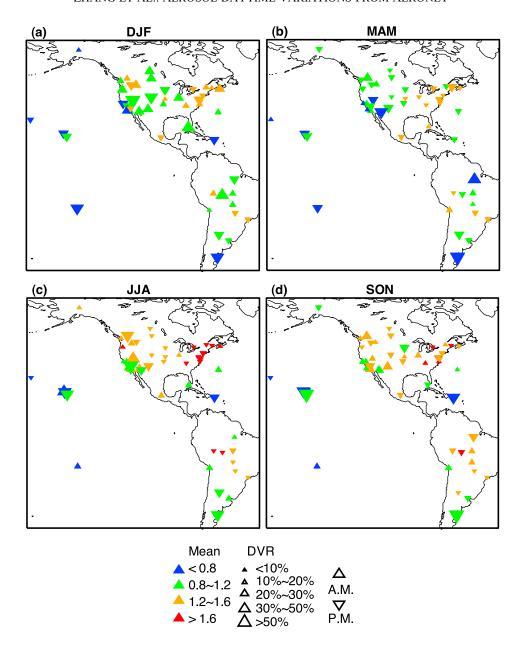


Figure 2. Same as Figure 1 but for aerosol Ångström exponent (AE) (over 440–870 nm).

than that in the East. Also the maximum AOD generally occurs in the morning in the West, which is opposite to that in the East. On the other hand, DVRs of AE in the West are significantly larger than that in the East in the summer and winter, suggesting that particle size or aerosol type in the West undergoes larger changes in the course of a day. However, uncertainties in computed AE are much larger at low AOD (given the AOD measurement uncertainty of ~ 0.01), therefore the larger DVR of AE in the West can be due at least in part to greater AOD uncertainties.

[15] Over South America, in the wet season (DJF and MAM), coarse mode biogenic aerosols from forests are a major component and some sites are also influenced by the long-range transport of Saharan dust and African smoke [Ansmann et al., 2009]. As such, AE in the wet season is relatively small, with a range of 0.8–1.2 for most sites but

less than 0.8 in some sites. In the wet-to-dry transition and dry season (JJA and SON), biomass burning smoke dominates over biogenic aerosols and AE is generally greater than 1.5.

[16] At island sites over the tropical Pacific Ocean and Atlantic Ocean where aerosol is dominated by marine aerosol with little influence from continental sources, both AOD and AE are generally smaller than that over the continents. However, the relative daytime variations of marine aerosol are generally large. For AOD, DVR generally falls into a range of 10~30% for all seasons, which however does not necessarily mean large absolute change of AOD because of small AOD values. For AE, the DVR is generally higher than 20%, with the highest value of more than 40% in Lanai in summer. While small AOD values over the ocean would have introduced large uncertainties in AE and its variations,

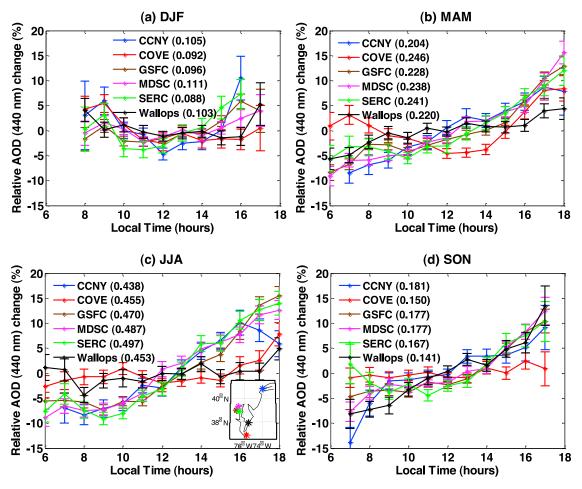


Figure 3. Percentage deviations of hourly average aerosol optical depth (AOD at 440 nm) relative to the daily mean in all seasons for sites over mid-Atlantic United States. The map in the lower right corner in JJA shows location of sites. The vertical bar represents the standard error of measurements in each hour. Seasonal mean AOD for each site are also shown in the figure.

the observed large daytime variations of AE are indeed consistent with some physical explanations to be discussed later.

[17] In the following sections, we examine in more detail the daytime variations of AOD and AE in several regions/sites representative of urban and industrial pollution, biomass burning smoke, marine aerosol, and free atmosphere aerosol.

4.2. Urban and Industrial Pollution Aerosols

4.2.1. Mid-Atlantic United States

[18] Several urban/suburban sites are located in the mid-Atlantic United States, including the Goddard Space Flight Center (GSFC), the Maryland Science Center in Baltimore (MDSC), City College of New York City (CCNY), the ocean platform of CERES Ocean Validation Experiment (COVE, off the coast of southern Virginia), Wallops Island (Virginia), and the Smithsonian Environmental Research Center (SERC, on the shore of the Chesapeake Bay in Maryland). As shown in Figure 3, these sites have comparable aerosol loading with high AOD (440 nm) of 0.44–0.50 in summer and low AOD of about 0.1 in winter. The day-time variation of AOD in spring and fall shows a pattern similar to but of lesser magnitude than that in summer. In summer, all the sites show similar patterns of daytime

AOD variation: a slight increase of AOD in the morning but a great increase of AOD in the afternoon. The DVR is about 20%, corresponding to AOD change of \sim 0.09 for GSFC, MDSC, CCNY, and SERC. For two coastal sites, COVE and Wallops, the DVR is less than 10%. Our results based on 12 years of observations from GSFC are consistent with that from an earlier study based on 1993-2001 measurements [Smirnov et al., 2002] These variations are likely associated with the photochemical production and hygroscopic growth as discussed in section 2. Because particles in the northeastern United States are mainly secondary sulfate aerosols [Bian et al., 2010] that are formed via photochemical and aqueous phase reactions [Malm, 1992], the increase of AOD over the daytime can be associated with photochemical processes, and at times also cloud processing. It is observed that the photochemical processes generally start in the early morning and persist about a half day [Sun et al., 2011].

[19] Given that sulfate is highly hygroscopic, a change of ambient RH over the day would contribute to the diurnal variation of AOD. The RH change over the day depends on altitude and location, as shown in Figure 4 for the six sites based on GEOS-4 assimilated meteorology in 2007 (similar daytime variations occur in 2006 and are not shown here).

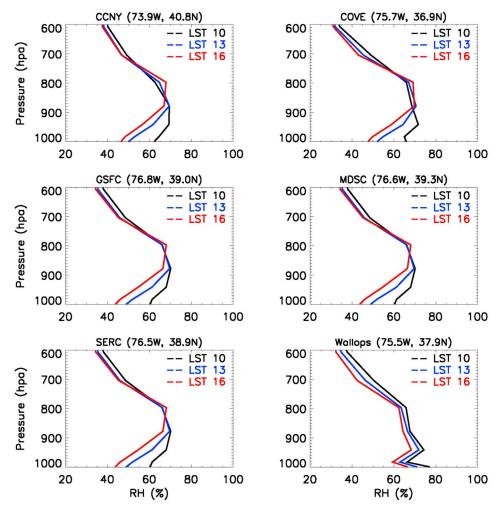


Figure 4. Diurnal relative humidity profiles from GEOS-4 at 10:00, 13:00, and 16:00 local standard time over mid-Atlantic sites in 2007.

Over Wallops, RH decreases from morning to afternoon at all altitudes and thus the AOD increase during the day cannot be explained by the hydroscopic growth. For the other five sites, the decrease of RH from morning to afternoon within the boundary layer would result in a decrease of AOD from morning to afternoon, which is however compensated by the increase of aerosol extinction due to the increase of RH near the top of the boundary layer. Although the RH increase near the top of the boundary layer is efficient in increasing the aerosol extinction because of the relatively high RH value, a majority of aerosols in the region stays within the boundary layer [e.g., Yu et al., 2010]. It is thus expected that the overall effect of RH change on AOD daytime variation may be relatively small. In winter, AOD is small and daytime variation range of AOD is <10% for most of those sites. High AOD in the morning and late afternoon in winter is consistent with the diurnal emission from local traffic. Unlike AOD, aerosol AE has a small daytime variation range of less than 10% (0.16) at all sites (not shown). RH could influence AE because hygroscopic aerosols can grow or evaporate with increasing or decreasing RH. However, the overall impact of RH on AE could be small because

an increase of size near the top of boundary layer is compensated by a decrease of size within the boundary layer.

4.2.2. United State West Coast

[20] Differing from the northeastern United States, the mean AOD at several California sites (Fresno, La Jolla, Monterey, and San Nicolas) show relatively small seasonal variations. AOD daytime variation in summer is also opposite to that in the northeastern United States, as shown in Figure 5a. AOD has its maximum in the morning and then decreases significantly until reaching a minimum in late afternoon, with DVR ranging from 20% to 38%. Correspondingly the absolute daytime change of AOD is 0.02~0.06, which is smaller than that at the northeastern sites, due to lower AOD not smaller DVR. This is qualitatively consistent with in situ measurements of aerosol concentrations in the region [Fine et al., 2004]. Similar daytime variation patterns are found in fall but with smaller magnitude. In winter and spring, no significant daytime variation is found (not shown). Such aerosol daytime variation is strongly controlled by the mesoscale circulations associated with unique topography in the region. For the three coastal sites in the Los Angeles basin and nearby, namely La Jolla,

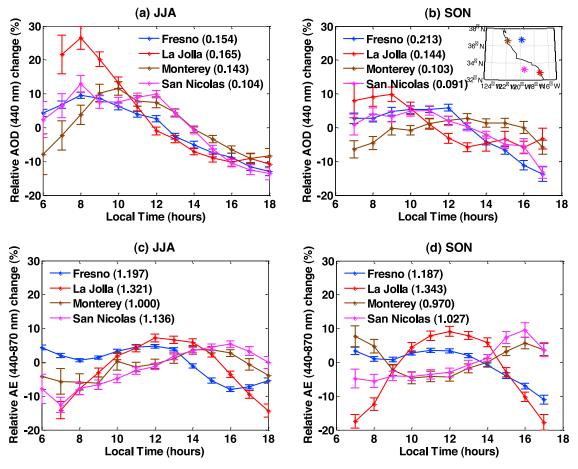


Figure 5. Percentage deviations of (a and b) hourly average aerosol optical depth (AOD at 440 nm) and (c and d) Angstrom exponent (AE) relative to their daily mean in JJA and SON for four sites over U.S. West Coast. The map in the upper right corner in Figure 5b illustrates locations of sites. The vertical bar represents the standard error of measurements in each hour. Seasonal mean of AOD/AE in each site are also shown in the figure.

Monterey, and San Nicolas (an island that is about 100 km offshore), the land-sea breeze circulations interacting with mountain ranges to the east of the basin control the evolution of aerosol [Cass and Shair, 1984; Wakimoto and McElroy, 1986; Lu and Turco, 1994, 1995]. At night the land breeze blowing toward the ocean assisted by mountain katabatic winds takes air pollutants from inland regions to the shore and offshore islands, resulting in an accumulation of pollutants in the coastal region [Cass and Shair, 1984]. This nighttime pollution accumulation, in combination with morning traffic, leads to a morning maximum AOD. With the development of the sea breeze during the day, relatively clean air from the ocean dilutes aerosol and lowers the AOD. Located further inland, Fresno is the second largest metro area in the Central Valley of California. There, AOD diurnal variation is closely related to the surface wind field pattern, especially in summer [Green et al., 1992]. Nighttime stable atmospheric stratification prevents the exit of air from the valley, causing accumulation of pollutants in the site. This in combination with morning traffic leads to a morning maximum AOD. During the day, a valley wind system develops with up-valley flow that ventilates pollutants out of the valley and reduces AOD. In addition, changes in relative humidity may also contribute to the aerosol daytime

variation. Daytime variation of AE could be 20–30% at La Jolla for both summer and fall, as shown in Figures 5c and 5d, respectively. The noontime peak AE may be associated with the decreasing relative humidity (decreasing particle size) and increasing photochemical activities (generating fine particles) from morning to noon and the dilution of small pollution particles with large marine particles as sea breeze brings in marine air in the afternoon.

4.2.3. Mexico City

[21] Aerosol daytime variations in Mexico City, one of the most polluted megacities in the world [Molina et al., 2007] are somewhat different from those in the northeast and western United States. As shown in Figure 6, seasonal mean AOD ranges from 0.38 in DJF to 0.51 in MAM; and seasonal mean AE is about 1.5 in all seasons, indicating the predominance of pollution aerosols throughout the year. Generally AOD increases from early morning, reaches maximum at noon or in early afternoon, and then more or less levels off. The daytime variation range of AOD is as large as 75% (corresponding to AOD change of 0.28) in DJF and 30–50% (corresponding to AOD change of 0.12–0.20) in other seasons, which is much stronger than that over the urban areas of the northeastern United States. The daytime

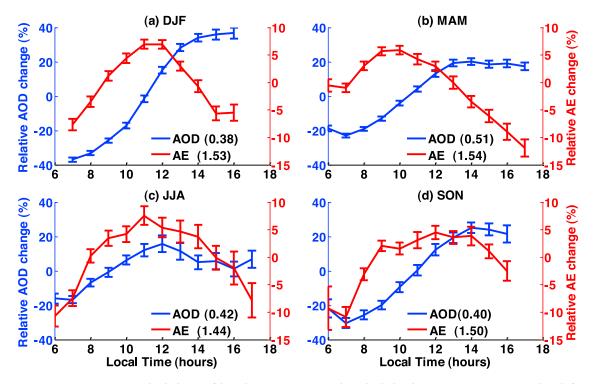


Figure 6. Percentage deviations of hourly average aerosol optical depth (AOD at 440 nm, using left y axis) and Ångström exponent (AE over 440–870 nm range, using right y axis) relative to the daily mean in four seasons in Mexico City. The vertical bar represents the standard error of measurements in each hour. Seasonal mean AOD and AE are also shown in the figure.

variation of AE is 10–15% with a peak in the late morning for all seasons.

[22] The daytime changes of AOD and AE in Mexico City are likely a combined effect of emission, photochemistry, and meteorological conditions associated with the complex topography. Mexico City is located within a basin confined on the east, south, and west sides by mountain ridges of about 1000 m in height with a broad opening to the north and the gap in the mountains at the southeast end of the basin. Local industrial and automobile emissions are two major sources of aerosol [Molina et al., 2007]. The precursor emissions of secondary organic aerosol (SOA) are higher in the morning than in the afternoon. SOA is efficiently formed shortly after sunrise [Molina et al., 2007]. In the morning, the city's unique topography and frequent atmospheric inversions trap the pollutants within the basin, likely leading to rapid increase of AOD throughout the morning [Whiteman et al., 2000; Fast et al., 2007]. In the afternoon, while the photochemical processes continue to produce aerosols, the basin is efficiently vented by terrain-induced winds. For example, the frequently developed strong southeasterly flow because of differential atmospheric heating [Raga et al., 1999; Doran and Zhong, 2000] brings in clean air from outside of the basin through the terrain gap in the southeastern corner and dilutes pollution in the city, resulting in the leveled off or slight decrease of AOD in the afternoon. Photochemical processes generate new particles, which are small in size, at late morning and noon [Salcedo et al., 2006] yielding a large AE. As the afternoon progresses those small particles are joined by large-size dust, kicked up by local winds, causing the AE to decrease.

4.3. Biomass Burning Aerosols in South America

[23] In the dry and dry-to-wet transition season (typically from August to October or ASO) of the central and southern Amazon, land clearing and pasture maintenance practices generate a large amount of carbonaceous aerosols [Andreae and Crutzen, 1997; Schafer et al., 2008]. Typically aerosol from biomass burning smoke accounts for ~90% of the fine particles and ~50% of the coarse particles [Martin et al., 2010]. Figure 7 shows daytime variations of AOD for four

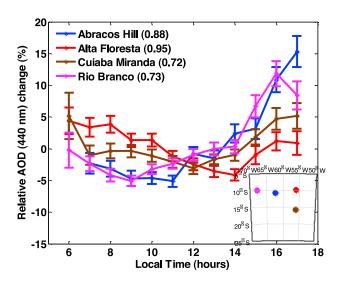


Figure 7. Same as Figure 3 but for sites over Amazon region during the dry season (August–October, ASO).

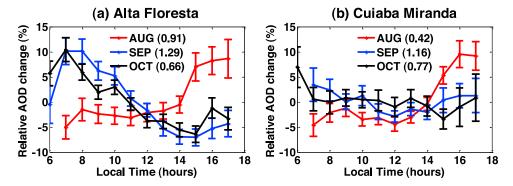


Figure 8. Percentage deviations of hourly average aerosol optical depth (AOD at 440 nm) relative to the daily mean for August, September, and October at Alta Floresta and Cuiaba-Miranda.

sites over the Amazon region, including Abracos Hill, Alta Floresta, Cuiaba-Miranda, and Rio Branco. AOD in all these sites show comparable seasonal mean AOD (0.72~0.96). A slight AOD decrease in the early morning and large increase in the afternoon have been observed for both Abracos Hill (about 15%) and Rio Branco (about 22%). On the other hand, in Alta Floresta and Cuiaba-Miranda, AOD generally shows both early morning and late afternoon peaks, with the minimum AOD around noon. The increase of AOD in the afternoon for all the sites is generally consistent with the documented occurrence of peak fire activities in the late morning and middle afternoon [Prins et al., 1998]. This diurnal trend in AOD is consistent with that observed in southern Africa in biomass burning regions of Zambia [Eck et al., 2003]. The AOD peaks in the early morning over Alta Floresta and Cuiaba-Miranda may have resulted from the long-range transport of smoke through the night, since both sites are usually influenced by both local biomass burning and long-range transport of aged smoke

[Prins et al., 1998; Reid et al., 1999]. Further analysis for Alta Floresta and Cuiaba-Miranda as shown in Figure 8 indicates that the AOD daytime variation changes with month. While the AOD daytime variation in September and October is similar to the seasonal average, AOD in August actually increases through the morning and peaks in late afternoon. These different daytime variations in different months are generally consistent with changing locations of biomass burning source regions with month. As discussed by Reid et al. [1999] these two sites are predominantly influenced by local pasture and grass fires in the early burning season but become more influenced by well-aged smoke transported from burning in the forest region in the late burning season (after mid-September).

4.4. Marine Aerosols

[24] In remote oceans where continental influences are minimal, aerosol is composed of sea salt and organics from sea spray, plus sulfate from dimethyl sulfide (DMS)

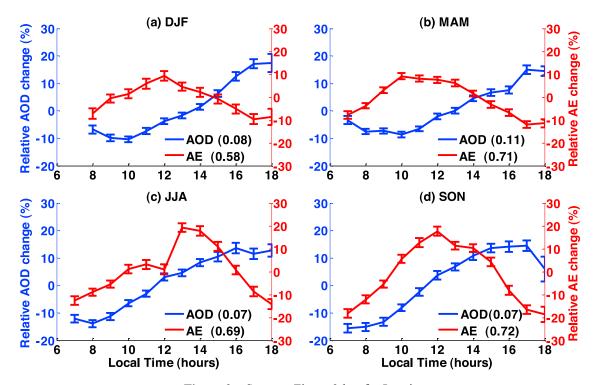


Figure 9. Same as Figure 3 but for Lanai.

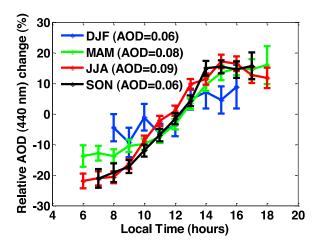


Figure 10. Deviations of hourly average aerosol optical depth (AOD at 440 nm) in four seasons at Table Mountain.

oxidations [Lewis and Schwartz, 2004]. While sea salt is dominated by coarse mode particles, sulfate and organic aerosol are fine mode. AERONET observations show that marine aerosol is bimodal, with a fine mode at effective radius of $0.11 \sim 0.14 \ \mu m$ and a coarse mode at $1.8 \sim 2.1 \ \mu m$ [Smirnov et al., 2003]. Figure 9 shows AOD and AE variations in Lanai, Hawaii. Lanai, with population of ~ 3000 , is mainly affected by marine aerosol with some influence from local pollution and springtime Asian pollution and dust, although episodic events of high-sulfate AOD occur from the emissions of nearby Kilauea and Mauna Loa volcanoes in Hawaii. AOD of 0.11 in spring is larger than 0.07–0.08 in other seasons, which is likely associated with springtime Asian transport [Eck et al., 2005]. The AOD daytime variation is similar in all seasons, with an early morning minimum and a late afternoon maximum. The daytime variation range is about 25%, corresponding to an AOD change of about 0.02. While 0.02 is comparable to the uncertainty of AERONET AOD measurements, the consistent daytime change shown by multiyear data might be indicative of physical processes. For example, the observed daytime variation may be linked to the alternation of wind direction between day and night. At night the island surface cools faster than surrounding ocean, which generates a wind from island toward ocean that cleans up the island. This may lead to a morning minimum AOD. During the day, the island warms faster than the ocean, resulting in a wind from ocean to island. This wind brings in marine aerosol and precursors (e.g., DMS) to island, which in combination with increasing photochemical activities could lead to a gradual increase of AOD during daytime. As shown in Figure 9, AE shows significant daytime variations with a peak around noon. The DVR for AE is about 10% in DJF and MAM but as much as 30% in JJA and SON. Although the large uncertainty of aerosol AE in such low AOD regime makes the detection of daytime variation difficult, the consistent daytime variation may indicate that active photochemistry produces fine mode sulfate aerosol and increases the AE around noon. However, aerosols on small islands, i.e., Midway Island and Bermuda, show a very small daytime variation with a very flat curve (figure not shown here). Because these two islands are tiny compare to Lanai, only a few kilometers in width and/or length,

aerosol properties on those islands remain close to the open ocean values.

4.5. Free Atmosphere Aerosol

[25] Table Mountain is located at an elevation of 2200 m in the San Gabriel Mountains, California, which is well above the ABL (about 1 km) in the Los Angeles basin [Wakimoto and McElroy, 1986]. It samples mainly free atmosphere aerosol but can be influenced by ABL aerosols in some occasions. As shown in Figure 10, the site has seasonal average AOD below 0.1 throughout the year. In DJF, the number of observations is too small to be used for detecting meaningful aerosol daytime variations. In other seasons, AOD consistently increases during the day and reaches a maximum in early afternoon. The AOD daytime variation range is about 25% in MAM and up to 35% in JJA and SON. The increase of AOD during the day is likely associated with the evolution of mountain-valley flows. After sunrise, the differential heating of atmosphere between the slope and nearby valley leads to upslope flows that could ventilate pollution from the Los Angeles basin upward to Table Mountain [Wakimoto and McElroy, 1986; Lu and Turco, 1995].

5. Concluding Remarks

[26] We have analyzed the daytime variations of aerosol optical depth and Angstrom exponent from 54 AERONET sites over the Americas and a few nearby islands on a seasonal basis. The analysis shows a wide range of AOD and AE daytime variations, depending on location and/or season. Mexico City shows the largest AOD in the afternoon, with a daytime variation range (DVR) at 440 nm of up to 75%. Such daytime changes of AOD are likely a combined effect of emissions and complex meteorology associated with the mountainous topography. In the mid-Atlantic United States several urban and suburban sites show consistently large DVR of AOD with the afternoon maximum, particularly in summer, which is likely associated with strong afternoon photochemical activities. On the other hand, several sites in the U.S. West Coast show relatively large DVR of AOD with the early morning maximum, possibly a combined effect of emission and topography-induced mesoscale circulations (such as land-sea breeze circulations and mountain-valley flows) and emissions. Similarly, the atmospheric boundary layer pollution aerosol can be transported upward by upslope flows associated with mountain-valley differential heating, resulting in an AOD increase throughout the day at high mountain sites. In the central part of the United States, aerosol daytime variations are generally small. Overall, human-influenced sites show a much larger daytime variation than natural sites.

[27] In Brazil, AOD in the burning season increases over the day with the late afternoon maximum consistent with observed peak biomass burning activities in midafternoon to late afternoon. However, in some sites AOD in the late burning season (late August to September) shows small daytime variation or even a morning maximum, which is likely associated with the increasing contribution of longrange transported smoke.

[28] Over islands of the remote Pacific Ocean with minimal influences from local anthropogenic activities and from

long-range transport of aerosols from upwind continents, AOD has an early morning minimum and a late afternoon maximum, which is presumably associated with land-sea breeze. Aerosol AE in the islands shows the largest value around noon, which may indicate an increase of fine mode non—sea-salt sulfate due to active photochemistry. For the open ocean, i.e., small islands like Midway and Bermuda, no obvious daytime variation of aerosol have been observed.

[29] In general, our study shows two typical daytime variations for AOD in a majority of AERONET sites: (1) AOD continuously increases during the day, reaching a maximum in the afternoon; (2) AOD peaks in the early morning but continuously decreases during the day. It appears that observations from polar orbiting satellites, such as Terra and Aqua, can't capture the maximum AOD, but may provide a good estimate of the daily average, although this would be by accident not by design since there are only two samples per day. To adequately capture the daytime variations, geostationary satellites such as the planned GEO-CAPE would need to make at least three successful aerosol retrievals during daytime in order to avoid aliasing. Given the often presence of clouds, satellites would need to be designed to sample at an hourly frequency.

[30] We have discussed some possible causes for the observed aerosol daytime variations based largely on previous studies of aerosol emissions, photochemical activity, and large-scale and mesoscale meteorology. Such discussion is generally qualitative in nature and not all variations are fully understood. To better understand the observed complex daytime variations, both comprehensive data sets and highresolution chemical transport model simulations are needed. Comprehensive data set of both aerosol and gaseous precursors at regional and continental scale with high temporal resolution cannot be obtained from low-Earth orbit but only from geostationary satellite missions. The unique capability of GEO-CAPE to simultaneously measure aerosol and its precursors would offer insights into how aerosol sources, chemical transformations, and transport processes determine the evolution of atmospheric aerosols on the hourly time scale.

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