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Impact of data quality and surface-to-column representativeness on the $PM_{2.5}$/satellite AOD relationship for the Continental United States

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Abstract

Satellite-derived aerosol optical depth (AOD) observations have been used to estimate particulate matter less than 2.5 µm ($PM_{2.5}$). However, such a relationship could be affected by the representativeness of satellite-derived AOD to surface aerosol particle mass concentration and satellite AOD data quality. Using purely measurement-based methods, we have explored the impacts of data quality and representativeness on the AOD inferred $PM_{2.5}$/AOD relationship for the Continental United States (CONUS). This is done through temporally and spatially collocated datasets of $PM_{2.5}$ and AOD retrievals from Aqua/Terra Moderate Resolution Imaging Spectroradiometer (MODIS), Multi-angle Imaging Spectroradiometer (MISR), and Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP). These analyses show that improving data quality of satellite AOD, such as done with data assimilation-grade retrievals, increases their correlation with $PM_{2.5.}$ However, overall correlation is relatively low across the CONUS. Also, integrated extinction observed within the 500 m above groud level (a.g.l.), as measured by CALIOP, is not well representative of the total column AOD. Surface aerosol in the Eastern CONUS is better correlated than in the Western CONUS. The best correlation values are found for estimated dry mass CALIOP extinction at 200–300 m a.g.l. and $PM_{2.5}$, but additional work is needed to address the ability of using actively sensed AOD as a proxy for $PM_{2.5}$ concentrations.

1 Introduction

Particulate matter (PM), especially suspended particles and solution droplets with diameters smaller than 2.5 µm ($PM_{2.5}$), contributes greatly to regional air pollution and can pose a threat to human health (e.g., Schwartz and Neas, 2000; Pope et al., 2002). Traditionally, the United States (US) Environmental Protection Agency (EPA) has monitored surface-based $PM_{2.5}$ concentrations using either a gravimetric-based method at ground stations with 24 h filter samplers or hourly Tapered Element Oscillat-

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ing Microbalance (TEOM) and beta gauge samplers (Federal Register, 1997). A number of studies have attempted estimates of surface-based $PM_{2.5}$ concentrations using satellite-retrieved aerosol optical depth (AOD) data (e.g., Hutchison, 2003; Wang and Christopher, 2003; Engel-Cox et al., 2004; Kumar et al., 2007; Liu et al., 2007; Hoff and Christopher, 2009). The advantages of estimating surface-based $PM_{2.5}$ concentrations using satellite-derived AOD data are obvious, as satellites, including both polar orbiting and geostationary satellites, typically provide a much larger spatial coverage than what can be inferred from ground stations over a broad surface footprint. However, data is limited to daylight cloud free conditions with once per day collection by polar orbiters (Diner et al., 1998; Remer et al., 2005) or multiple images in morning or afternoon from geostationary satellites (Zhang et al., 2001; Prados et al., 2007).

Previous research efforts have focused on algorithm development for solving PM proxies based on AOD. For example, Chu et al. (2003) compare PM_{10} concentrations with surface AOD measurements from the Aerosol Robotic Network (AERONET) in northern Italy and highlight the potential of using Moderate Resolution Imaging Spectroradiometer (MODIS; Remer et al., 2005) AOD as an estimate for PM_{10} concentration. Several studies have focused on correlating satellite AOD observations and $PM_{2.5}$ concentrations (e.g., Wang and Christopher, 2003; Liu et al., 2004), and advances have been made improving correlation between the two by considering other meteorological and environmental parameters, such as the surface mixed-layer height (Engel-Cox et al., 2006; Gupta et al., 2006) and relative humidity (Shinozuka et al., 2007; Van Donkelaar et al., 2010). Simulated vertical structure from chemical transport models (e.g., Van Donkelaar et al., 2006, 2010) has also been used to help improve the $PM_{2.5}$/satellite AOD relationship.

There are important issues, however, that need be considered when applying satellite-based observations in general, much less as a proxy for $PM_{2.5}$ estimates. First, uncertainties exist in satellite-retrieved AOD values due to issues such as cloud contamination, inaccurate optical models used in the retrieval process and heterogeneous surface boundary conditions (e.g., Zhang and Reid, 2006; Shi et al., 2011a;

Toth et al., 2013). Even today, convergence has not yet been reached for retrieved AOD values found among the most widely used satellite aerosol products, such as the Dark Target/DeepBlue MODIS and Multi-angle Imaging Spectroradiometer (MISR; Diner et al., 1998; Kahn et al., 2010) aerosol products (e.g., Shi et al., 2011b). Any estimate of $PM_{2.5}$ derived from satellite AOD data cannot be more accurate than the AOD data themselves. Thus, relationships between AOD and $PM_{2.5}$ are likely to be highly sensor product specific. Second, AOD derived from passive sensors is a column-integrated value, and $PM_{2.5}$ concentration is a surface measurement. Under conditions where aerosol particles are concentrated primarily within the surface/boundary layer, AOD is presumably a likelier proxy for $PM_{2.5}$ concentration. Conversely, in conditions where aerosol plumes are transported above the boundary layer, AOD will likely prove a weaker one. Finally, AOD is a column-integrated sum of total particle extinction, whereas $PM_{2.5}$ is measured with respect to dried particle ingested for analysis by corresponding instruments. Thus, hygroscopicity and mass extinction efficacy corrections are further required to accurately characterize any relationship present between the two parameters.

While some studies have attempted to use chemical transport models and ground-based lidars to investigate a relationship between aerosol particle structure, column-integrated AOD and surface-based $PM_{2.5}$ (Liu et al., 2004; Van Donkelaar et al., 2006; Boyouk et al., 2010; Hyer and Chew, 2010), a measurement-based analysis using the Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP; Winker et al., 2007; Hunt et al., 2009) would allow for such a study over relatively-broad spatial and temporal scales, for which more tenable proxies between AOD and $PM_{2.5}$ may be realized and thus applied on more representative scales. Range-resolved information collected with CALIOP provides the critical perspective for relating the depth and vertical extent of aerosol particle presence to both surface-based $PM_{2.5}$ measurements and passive retrievals of column-integrated AOD.

In this study, we examine the $PM_{2.5}$/AOD relationship for the Continental United States (CONUS) from the unique joint perspectives of data quality and aerosol surface-

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to-column representativeness. Through the use of MODIS, MISR, and CALIOP observations, the following research questions are considered:

1. How does the quality of passive satellite AOD retrievals impact the $PM_{2.5}$/AOD relationship?

2. Based on CALIOP data, how representative are surface-based measurements to aerosol particle presence within the full column?

3. Can near surface observations from CALIOP be used as a better proxy for $PM_{2.5}$ concentration?

The paper has been designed to discuss each component sequentially, thus building off the previous step. In Sect. 2 of this paper, we describe the various satellite and surface-based datasets used. In Sect. 3, the $PM_{2.5}$/AOD relationship is first examined at an hourly timescale, followed by a daily analysis in which we explore the impact of AOD quality on this relationship. In Sect. 4, we investigate the representativeness of satellite-derived surface aerosol concentration to that of the entire column, and how well surface AOD correlates with total column AOD. Lastly in Sect. 5, we provide results comparing surface-based $PM_{2.5}$ and CALIOP aerosol extinction near the lower bounds of the satellite profile to investigate the potential use of CALIOP data for air quality applications.

2 Datasets

2.1 MODIS, MISR, and CALIOP Data

Aboard both the NASA Aqua and Terra satellites, MODIS is a spectroradiometer with 36 channels (0.41 to 15 µm), seven of which (0.47 to 2.13 µm) are applied operationally for the retrieval of aerosol particle optical properties. The Dark Target Level 2 products created from these retrievals are reported at a spatial resolution of $10 \times 10\,km^2$, with over-land uncertainties of $0.05 \pm 0.15 \cdot$ AOD (Remer et al., 2005). This study utilizes the Corrected_Optical_Depth_Land (0.550 µm) parameter of Dark Target Level 2 Collection 5.1 retrievals from Aqua (MYD04_L2) and Terra (MOD04) MODIS (2008–2009, operational), with quality assurance (QA) limiting the analysis to only those retrievals with Quality_Assurance_Land parameter flags of "very good". Although the DeepBlue (DB) MODIS aerosol products also provide aerosol retrievals over land, the Collection 5.1 Aqua DB MODIS aerosol products are not available for the study period and are thus not included in the study.

MISR, aboard the Terra satellite, is a unique spectroradiometer, able to collect observations at nine different viewing angles, providing a means for studying aerosol particle size and shape (Diner et al., 1998). MISR features four spectral bands, located at 0.446, 0.558, 0.672, and 0.867 µm. Different from the Dark Target MODIS aerosol products, the MISR aerosol product also includes AOD retrievals over bright surfaces such as desert regions. Kahn et al. (2005) suggested that 70 % of MISR AOD data are within 0.05 (or 20 % × AOD) of sun-photometer measured AOD values. This study utilizes the same two years (2008–2009) of AOD derived from Version 22 MISR retrievals (0.558 µm), flagged through QA screening as "successful".

CALIOP is a multi-wavelength (0.532 and 1.064 µm) polarization lidar flown aboard the Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observations (CALIPSO) platform within the NASA "A-Train" constellation (e.g., Stephens et al., 2002). To gain an understanding of aerosol particle distribution over the US for 2008–2009, this study utilizes the Version 3.01 CALIOP Level 2 5 km Aerosol Profile (L2_05kmAProf) (Winker et al., 2007, 2012) product. The Version 3.01 Level 2 Vertical Feature Mask (L2_VFM) product is also used to restrict the analysis to those 5 km AOD and total extinction (at 0.532 µm) profile retrievals that are cloud-free, in a manner consistent with that of Toth et al. (2013).

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2.2 Quality-assured MODIS and MISR Subsets

Existing uncertainties in passive satellite AOD retrievals, such as those for MODIS and MISR, are optimally suppressed before being considered and applied for data assimilation (DA) activities involving operational aerosol forecast models (e.g., Zhang et al., 2008). Through rigid QA, reduced AOD uncertainties have been characterized and DA-quality AOD datasets have been created for both over land (Hyer et al., 2011) and over ocean MODIS DT products (Shi et al., 2011a), as well as the MISR aerosol products (Shi et al., 2011b, 2012). In this study, we use DA-quality MODIS and MISR AOD products as control datasets for comparison with operational MODIS and MISR products.

Available at 6 hourly 1° × 1° resolution, DA-quality AOD data are converted to daily averages and then compared with daily $PM_{2.5}$ concentrations. For comparison purposes with the $PM_{2.5}$ data available (described further below), we have constructed daily-averaged "Level 3" AOD data using operational MODIS and MISR aerosol products after applying first-order QA as described in Sect. 2.1. DA-quality MODIS aerosol products are available from the Global Ocean Data Assimilation Experiment (GODAE) server (http://www.usgodae.org/). However, no quality-assured hourly DA-quality aerosol products are currently available, and no comparisons were therefore made between the DA-quality products and hourly $PM_{2.5}$ measurements.

2.3 Surface $PM_{2.5}$

The US EPA has collected observations of surface-based PM since the passage of the Clean Air Act in 1970 (http://www.epa.gov/air/caa/). In 1997, the EPA began specifically monitoring $PM_{2.5}$ concentrations (Federal Register, 2006). The Federal Reference Method (FRM), a filter-based method, is used to measure concentration over a continuous 24 h period. The filter is weighed before and after the sample collection interval and $PM_{2.5}$ mass concentration ($\mu g\,m^{-3}$) is calculated by dividing the total mass of $PM_{2.5}$ particles by the volume of air sampled (Federal Register, 1997). Some EPA

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sites also report hourly (continuous) $PM_{2.5}$ measurements. For this study, two years (2008–2009) of daily and hourly $PM_{2.5}$ Local Conditions (EPA Parameter Code 88101) data were used and obtained from the EPA Air Quality System (AQS).

3 How does the quality of passive satellite AOD retrievals impact their linear correlation with surface-based $PM_{2.5}$?

As a first step, linear correlations between passive satellite AOD retrievals and $PM_{2.5}$ observations in the United States are derived. We investigate the impact of data quality to the AOD/$PM_{2.5}$ relationship through a daily analysis using both daily-averaged operational and DA-Quality AOD datasets, as well as daily $PM_{2.5}$ data. No hourly DA-quality AOD retrievals are currently available, and therefore the impact of data quality to the AOD/$PM_{2.5}$ correlations are not specifically characterized on this temporal scale. Still, an hourly analysis is first considered, using only operational AOD data and hourly $PM_{2.5}$ data, for comparison purposes and for establishing a relevant context for the relationship between AOD and $PM_{2.5}$.

Figure 1 depicts those $PM_{2.5}$ monitoring sites for the 2008–2009 period that reported hourly (Fig. 1a) and daily-averaged (Fig. 1b) $PM_{2.5}$ observations. A total of 102 sites reported hourly data, while 991 sites collected daily data (see figure caption for color scheme). Note that some sites feature multiple instruments observing $PM_{2.5}$ concentration; one routine/primary, regular measurement and a secondary measurement that is only available sporadically. Both types of $PM_{2.5}$ data are included for this analysis.

3.1 Hourly analysis

For 2008–2009, the operational Level-2 AOD datasets are spatially and temporally collocated with available $PM_{2.5}$ observations. After these AOD data are filtered through basic QA screening (Sect. 2.1), each hourly $PM_{2.5}$ observation is matched with those Level-2 AOD retrievals meeting the QA criteria and found within 40 km and 1 h of the

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$PM_{2.5}$ observation. All remaining AOD values are then averaged for a single comparison with the $PM_{2.5}$ observation. We chose 40 km as the averaging range for the satellite data after assuming a mean wind speed of $10\,m\,s^{-1}$ influencing aerosol plumes transport (approximately $40\,km\,h^{-1}$). AOD autocorrelation at or exceeding 0.8 has been reported for a distance of 40 km (on average) (Anderson et al., 2003; Zhang et al., 2011), making this a reasonable constraint.

Table 1 summarizes the results of the hourly collocation of $40\,km\,h^{-1}$ average MODIS/MISR AOD with corresponding ground-based $PM_{2.5}$ measurements over the two year study, including linear correlation coefficients and data counts for the contiguous US divided into its four respective time zones: Eastern (UTC-5), Central (UTC-6), Mountain (UTC-7), and Pacific (UTC-8). Relatively low correlations are found for the US, as a whole. However, a regional dependence of the relationship between the two parameters is also apparent. The Eastern US region exhibits higher correlation than does the Pacific US by a factor of nearly two (0.2 vs. 0.4). This is consistent with several studies that have shown similar regional effects. For example, Hu (2009) reports average $PM_{2.5}$/AOD correlations of 0.67 (Eastern US) and 0.22 (Western US), with Engel-Cox et al. (2004) and Paciorek et al. (2008) reporting similar correlations of 0.6–0.8 (Eastern US) and 0.2–0.4 (Western US). It has been suggested that this regional variability in the $PM_{2.5}$/AOD relationship is due to differences in topography, surface albedo, and boundary layer depth between the Eastern and Western US (Engel-Cox et al., 2006).

In Fig. 2, regional differences of $PM_{2.5}$/AOD correlation are also evident from scatterplots for the Eastern (Fig. 2a) and Pacific (Fig. 2b) time zones, with greater linearity observed in the Eastern US compared to the west. Also, $PM_{2.5}$ concentration averages were computed for each 0.1 bin of AOD, and shown with respect to both Terra MODIS and MISR. Note that although we have listed both Aqua and Terra MODIS in Table 1, we show only the Terra MODIS/MISR analysis in Fig. 2 because of their common satellite-observing platform. In general, a better correlation is found for the bin averages, which is consistent with that reported by Gupta et al. (2006).

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Seasonally, each of the hourly $PM_{2.5}$/AOD correlations coefficients shown in Table 1 are recomputed for December through May (Table 1; DJFMAM) and June through November (Table 1; JJASON). There are fewer data points for DJFMAM than JJASON (∼ 68 % decrease), enhanced by the absence of December 2007 in the dataset. Overall, however, lower correlations are found during this season compared with the annual mean. The opposite is thus true for JJASON. Although not shown here, further analysis reveals that higher correlations of JJASON may be due to a significant number of cases of relatively high $PM_{2.5}$ (greater than 35 $\mu g\,m^{-3}$) and high satellite AOD (greater than 0.3) that occur during this season, relative to DJFMAM, which may positively influence the regression compared with JJASON.

3.2 Daily analysis

We next investigate how the relationship between AOD and $PM_{2.5}$ is affected by the perceived data quality of the operational satellite AOD datasets, using only basic QA, vs. the DA-quality Level 3 AOD data. As discussed above, these latter data are subject to more advanced screening, with filtering, correction, and spatial aggregation applied. Each available daily ground-based $PM_{2.5}$ observation is matched with both the operational and DA-quality AOD retrievals found within 1° latitude/longitude and the day of the $PM_{2.5}$ observation. Results of the daily 1° × 1° operational and DA-quality MODIS/MISR AOD analyses are shown for the CONUS and each respective time zone in Table 2.

Distinct increases are found for $PM_{2.5}$/AOD correlation using the DA-quality satellite AOD products vs. the operational satellite AOD datasets (Table 2). For example, $PM_{2.5}$/AOD correlations for the CONUS increase by about 0.12 (Aqua MODIS), 0.16 (Terra MODIS), and 0.14 (MISR) from each respective operational to DA-quality dataset. Note that data counts for each DA-quality AOD analysis decrease relative to each corresponding operational AOD analysis, indicative of fewer available collocations from the Level 3 AOD datasets from increased data rejection. We believe that such a pronounced pattern reflects the influence of AOD retrieval quality from the passive satellites on their relationship with surface-based $PM_{2.5}$ measurements.

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Also shown in Table 2, the Eastern sample exhibits greater linearity (i.e., correlation) overall compared with the Western one. Figure 3 further illustrates the regional variation in $PM_{2.5}$/DA AOD correlation, through corresponding scatterplots for the Eastern (Fig. 3a) and Pacific (Fig. 3b) time zones. As in Fig. 2, we only show the Terra MODIS/MISR analysis because of their common platform. Also, averages of $PM_{2.5}$ concentrations are shown for each 0.1 bin of DA TERRA and MISR AOD.

The seasonality of the $PM_{2.5}$/AOD relationship for the daily analysis is investigated in Table 2. As encountered above for Table 1, there are fewer data points for DJFMAM than JJASON ($\sim 32\,\%$ decrease). Likewise, lower $PM_{2.5}$/AOD correlations are found during DJFMAM, and higher correlations are found from JJASON, as compared to the mean annual results presented in Table 2. Again, this pattern may be due to a larger number of high $PM_{2.5}$ (greater than $35\,\mu g\,m^{-3}$) and high satellite AOD (greater than 0.3) values that are found from JJASON, as compared to DJFMAM. However, a longer study period is likely needed to more appropriately understand the seasonal dependence of the $PM_{2.5}$/AOD relationship.

Figure 4 consists of two maps depicting daily $PM_{2.5}$ sites used in this analysis, color-coded with respect to $PM_{2.5}$/AOD correlation coefficient. Figure 4a reflects the $PM_{2.5}$/daily operational Terra MODIS AOD relationship, with generally higher correlations in the Eastern US than the Pacific US. Figure 4b illustrates a clear increase in $PM_{2.5}$/AOD correlation for the daily DA Terra MODIS AOD analysis, with again still higher correlations for the Eastern US compared to those results found in the west. Similar regional and operational-to-DA AOD patterns in the $PM_{2.5}$/AOD relationship are shown in Fig. 5 for the operational MISR AOD (Fig. 5a) and DA MISR AOD (Fig. 5b) daily analyses.

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4 How representative is the surface layer aerosol particle presence to the atmospheric column?

We have demonstrated that the quality of the AOD datasets investigated impacts any linear correlation apparent with ground-based $PM_{2.5}$ measurements. Next we explore the representativeness of aerosol particle presence near the surface to that of the atmospheric column. We use the CALIOP L2_05kmAProf product, featuring a vertical resolution of 60 m for altitudes below 20.2 km above mean sea level (a.m.s.l.). Using the corresponding mean surface elevation reported with each profile, values of extinction coefficient and AOD (0.532 µm) are re-gridded linearly at 100 m resolution vertically from the surface (above ground level, or a.g.l.) to 8.2 km after a robust QA screening procedure takes place. The details of this QA process are documented in past studies (Kittaka et al., 2011; Campbell et al., 2012a; Winker et al., 2012; Toth et al., 2013). Only cloud-free profiles are considered.

Shown in Fig. 6 are $1^\circ \times 1^\circ$ averages (relative to the number of cloud free 5 km CALIOP profiles in each $1^\circ \times 1^\circ$ regional bin) of 0.532 µm aerosol extinction coefficient for the 0.0 to 0.5 km layer (Fig. 6a), 0.5–1.5 km (Fig. 6b), 1.5–2.5 km (Fig. 6c) and 2.5–3.5 km a.g.l. (Fig. 6d), respectively. In general, extinction values observed in the lower atmospheric layers (Fig. 6a and b) are larger than those observed in the elevated atmospheric layers (Fig. 6c and d). However, higher mean values are found nearer the surface in the eastern region (particularly the southeastern US; Fig. 6a and b), while higher values are found at elevated heights in the west (Fig. 6c and d). These data indicate that, on average, aerosol particle distributions tend to be more concentrated near the surface in the east and more diffuse vertically in the west.

Corresponding with Fig. 6a, Fig. 7 is a plot of the average percentage of surface layer-integrated extinction (altitudes lower than 500 m a.g.l.) to total column AOD. We use the average of the lower 500 m a.g.l. to represent the surface layer so as to minimize ground flash contamination in the CALIOP data when observations are near the ground (e.g., Campbell et al., 2012b). Values are generally below 40 % across the

CONUS, with higher values more concentrated in the eastern part of the country. The distribution is noisy, however, and thus to better interpret these data, we present a five-year assessment (2006–2011) of CALIOP data (Fig. 8). Common patterns emerge, though more distinctly, as higher percentages are again found over the east vs. the west. In general, however, AOD below 500 m a.g.l. accounts for only 30 % or less of the total column AOD across the US. This indicates that it is necessary to have a priori knowledge of the ratio between near-surface integrated extinction to column-integrated AOD in order to better characterize the likely representativeness of applying satellite AOD as a proxy for surface $PM_{2.5}$ concentration.

Note that although integrated extinction over the lowest 500 m a.g.l. may not be representative of the total column AOD, it is possible that the correlation between the two could be high, and thus useful for satellite AOD/$PM_{2.5}$ studies. Although not shown here, we also compute the $1^\circ \times 1^\circ$ average correlation between integrated extinction from the lowest 500 m a.g.l. and the total column AOD. Globally over land, an average correlation of 0.61 is found. For the United States, a similar value of 0.62 is calculated, with values of 0.61 for the Eastern time zone and 0.57 for the Pacific. Importantly, the lack of significant regional variability in these relationships indicates that although the Eastern and Pacific time zones may exhibit different AOD surface contribution percentages, integrated surface extinction correlates relatively consistently with total column AOD. Still, given a perfect possible correlation of 1 between integrated surface level extinction and $PM_{2.5}$ concentration, the correlation value of ~ 0.6 between the former with column-integrated AOD might represent the best case scenario, on a regional average, that one could derive presently for the satellite AOD to $PM_{2.5}$ concentration relationship.

To evaluate the influence of aerosol particle presence at elevated levels, in Fig. 9a we show the fraction of CALIOP-retrieved column-integrated AOD found above an arbitrary standard height of 2 km a.g.l., thus segregating mostly boundary layer particle presence vs. those propagating within the free troposphere. It is evident that regional variations in the fraction of AOD above 2 km exist, as the western half of the US ex-

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hibits at least double the amount of particle extinction above 2 km than does the eastern US. However, note that many areas in California, where a relatively dense array of Pacific US $PM_{2.5}$ sites are located, exhibit relatively low contributions comparable to that of the east (usually below 30 %). Consistent with the findings shown in Fig. 9a, regional variations in the frequency of occurrence of AOD above 2 km a.g.l. are also observed (Fig. 9b), with generally higher frequencies in the west as compared to the east. While the average frequency of occurrence of AOD above 2 km a.g.l. for the US is ∼ 40 % (Fig. 9b), a value of ∼ 20 % (not shown) is computed for the frequency of occurrence of when at least 50 % of aerosol (as measured by CALIOP AOD) within an atmospheric column is found above 2 km a.g.l. This indicates a significant number of elevated aerosol plumes occurred over the US during the 2008–2009 period, and thus will not be recognized by surface-based $PM_{2.5}$ measurements.

5 Can near surface observations from CALIOP be used as a better proxy for $PM_{2.5}$ concentration?

Taking advantage of an active-profiling aerosol particle sensor like CALIOP, we investigate the relationship between hourly $PM_{2.5}$ concentration and CALIOP 0.532 µm extinction coefficient values near the surface. The temporal/spatial collocation and 40 km AOD averaging process here is the same as described in Sect. 3. Recall that $PM_{2.5}$ is a dry particle mass measurement. However, satellite-retrieved AOD values include the effects of aerosol particle growth as a function of vapor pressure. To compute the CALIOP extinction and $PM_{2.5}$ relationship, a sensitivity study was performed for which the hygroscopic growth of aerosol particles was accounted for. We approximate that aerosol particles over the US are sulfate aerosols, and apply the sulfate aerosol hygroscopic growth factor (Hanel, 1976; Hegg et al., 1993; Anderson et al., 1994) to compute dry aerosol extinction and AOD using Goddard Modeling and Assimilation Office (GMAO) relative humidity values included as metadata in the NASA-disseminated CALIOP files. No correction is made to extinction coefficient values when relative hu-

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midity is less than 30 % or above 95 %. Further, we investigate the sensitivity of the CALIOP value chosen to compare with by varying the height of the retrieval used between 0 and 500 m a.g.l. in 100 m segments.

Results, including the level of CALIOP extinction used, are summarized in Table 3. For both the Eastern and Pacific US time zones, altering the level of the reported CALIOP extinction from 200 to 500 m a.g.l. has little effect on correlation. Relatively low correlation is observed using the CALIOP extinction values at the 0–100 m level, however, suggesting the likely impacts of ground contamination of the backscatter signal. When hygroscopic growth of aerosol particles is considered, modest improvements are found for the Eastern US but not the climatologically drier Pacific region.

We next investigate the relationship between CALIOP extinction near the surface and $PM_{2.5}$ concentrations when collocated Aqua MODIS operational retrievals are available. This $PM_{2.5}$/CALIOP/Aqua MODIS dataset was constructed for both hourly and daily analyses during the 2008–2009 period. For the hourly study, both CALIOP and operational Aqua MODIS observations are again averaged within 40 km and the 1 h of the $PM_{2.5}$ measurements. For the daily comparison, observations from CALIOP are averaged within 100 km along-track (approximately 1°), and those from operational Aqua MODIS are averaged within 1° latitude/longitude, and the day of each $PM_{2.5}$ measurement.

Figure 10 shows hourly analysis results for dry mass-adjusted CALIOP extinction at 200–300 m a.g.l. (Fig. 10a) and operational Aqua MODIS AOD (Fig. 10b). The 200–300 m layer was used because the lowest 200 m a.g.l. of retrieved extinction is considered subject to ground contamination (e.g., Schuster et al., 2012; Omar et al., 2013). Reasonably high correlations of ~0.8 are found for CALIOP/$PM_{2.5}$ for both the Eastern and Pacific time zones. A difference exists between these two regions for Aqua MODIS, however. The Eastern US exhibits similar correlation compared with that found above from CALIOP, but drops off to about ~0.5 for the Pacific US. Clearly, CALIOP and Aqua MODIS retrievals behave similarly for the Eastern US, but CALIOP performance is much better than Aqua MODIS over the Pacific.

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Figures 11a and b depict the same analyses as in Fig. 10, but now for the daily analysis of $PM_{2.5}$/CALIOP/Aqua MODIS. Correlations are reduced for each time zone, compared with the hourly results. As was shown in Fig. 10, CALIOP and Aqua MODIS exhibit similar correlations with daily $PM_{2.5}$ for the Eastern US, but daily $PM_{2.5}$/CALIOP correlations are better than daily $PM_{2.5}$/Aqua MODIS correlations for the Pacific US.

CALIOP near-surface extinction/hourly $PM_{2.5}$ relationships represent the most consistent correlations solved in this study. However, more research is necessary to advance our understanding of the relationship between actively-profiled aerosol optical properties and $PM_{2.5.}$ This is particularly important since studies have reported significant uncertainties in CALIOP AOD and extinction data (e.g., Schuster et al., 2012; Omar et al., 2013), especially for values lower than 200 m a.g.l., which are clearly critical to resolving the most optimal CALIOP extinction/$PM_{2.5}$ relationship. Note, however, that aside from ground contamination issues described above, Campbell et al. (2012a, b) argue for an additional QA step of removing CALIOP profiles from bulk averages where no aerosol extinction is retrieved below 200 m to limit the effects of signal pulse attenuation. This effect may be further contributing to lower skill at these heights. Further, additional analysis can be further explored where the top height of the surface-detached mixed aerosol layer is known. This constraint was not considered here, and is outside the general scope of our investigation.

6 Conclusions

Surface measurements of particulate matter with diameters less than 2.5 µm ($PM_{2.5}$) are a frequent tool used to evaluate air quality in urban areas. Past studies have investigated the ability of using aerosol optical depth (AOD) retrievals from passive satellite sensors as proxies for $PM_{2.5}$ concentrations. Extending from past efforts, this study explores the impact of passive satellite AOD data quality and satellite-derived surface-to-column aerosol representativeness on the $PM_{2.5}$/AOD relationship for a two-year period (2008–2009). With a focus on the United States, passive AOD operational Level-

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2 retrievals from Aqua/Terra Collection 5.1 Moderate Resolution Imaging Spectroradiometer (MODIS) and Version 22 Multi-angle Imaging Spectroradiometer (MISR) are temporally and spatially collocated for an hourly comparison with $PM_{2.5}$ measurements. Next, operational and data assimilation (DA) quality Aqua/Terra MODIS and MISR AOD datasets are analyzed against $PM_{2.5}$ on a daily temporal scale to reveal the effects that AOD data quality can exhibit with respect to $PM_{2.5}$/AOD correlations. The representativeness of surface aerosol particle concentration to that of the entire column, as well as the correlation between surface AOD and total column AOD, are investigated using observations from Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP). CALIOP is then used to examine the relationship between near surface aerosol extinction and $PM_{2.5}$.

The conclusions of this study are summarized as follows:

1. Application of aggressive QA procedures to passive satellite AOD retrievals increases their correlation with $PM_{2.5}$ for all of the CONUS.

2. Correlations remain low even with aggressive QA.

3. Near-surface extinction (below 500 m a.g.l.), as measured by CALIOP, is not well representative of total column-integrated extinction (i.e., AOD). Regionally, near-surface aerosols are more representative of total column AOD in the Eastern US than in the Western US.

4. Correlations between near-surface CALIOP 0.532 µm extinction and hourly $PM_{2.5}$ observations are better than can be achieved with passive AOD retrievals. However, with fewer than 100 pairs of collocated $PM_{2.5}$ and CALIOP extinction data points used, such a finding is tenuous. Additional studies are needed to further explore the possibility of accurately estimating $PM_{2.5}$ concentrations from surface extinction derived from active sensors.

In this paper, we have demonstrated that estimation of $PM_{2.5}$ concentrations from satellite retrieved AOD is limited by both the quality of satellite AOD retrievals as well as the representativeness of column-integrated AOD to near surface AOD. The use of near surface extinction measurements from active sensors, such as CALIOP, may provide a better $PM_{2.5}$ relationship for operational estimates. However, ground contamination for near-surface CALIOP measurements and the effects of humidity on aerosol optical properties need further investigation. Still, satellite derived aerosol properties are of much value to $PM_{2.5}$ studies, especially with the synergistic use of passive and active aerosol-sensitive observations, and through assimilating these quality-assured data into air-quality focused numerical models for future $PM_{2.5}$ monitoring and forecasts.

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Table 1. Correlation coefficients and data counts of the 40 km average operational Aqua/Terra MODIS and MISR AOD/hourly $PM_{2.5}$ collocation analyses for the Eastern, Central, Mountain, and Pacific time zones and Continental United States total for the entire two-year (2008–2009) study period, December through May 2008–2009 (DJFMAM), and June through November 2008–2009 (JJASON).

Dataset		Operational Aqua MODIS		Operational Terra MODIS		Operational MISR	
		R value	Data Count	*R* value	Data count	*R* value	Data count
Eastern	All	0.57	2081	0.47	2748	0.42	614
	DJFMAM	0.49	477	0.39	566	0.11	154
	JJASON	0.57	1551	0.50	2001	0.50	408
Central	All	0.27	1765	0.22	2005	0.22	447
	DJFMAM	0.11	335	0.14	346	0.16	112
	JJASON	0.38	1330	0.28	1511	0.26	304
Mountain	All	0.19	1369	0.12	1632	0.10	391
	DJFMAM	−0.08	215	0.09	250	0.16	95
	JJASON	0.30	1136	0.17	1354	0.20	277
Pacific	All	0.15	3832	0.22	3873	0.11	903
	DJFMAM	0.08	1064	0.21	1047	0.15	269
	JJASON	0.26	2560	0.21	2564	0.29	539
Contiguous US	All	0.19	9047	0.22	10 258	0.15	2355
	DJFMAM	0.03	2091	0.12	2209	0.07	630
	JJASON	0.34	6577	0.25	7430	0.27	1528

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Table 2. Correlation coefficients and data counts of the daily 1° × 1° average operational DA Aqua/Terra MODIS and MISR AOD/daily $PM_{2.5}$ collocation analyses for the Eastern, Central, Mountain, and Pacific time zones and Continental United States total for the entire two-year (2008–2009) study period, December through May 2008–2009 (DJFMAM), and June through November 2008–2009 (JJASON).

Dataset		Aqua MODIS				Terra MODIS				MISR			
		Operational		DA		Operational		DA		Operational		DA	
		R value	Data Count	*R* value	Data Count	*R* value	Data Count	*R* value	Data Count	*R* value	Data Count	*R* value	Data Count
Eastern	All	0.40	76 194	0.50	29 682	0.38	80 810	0.51	38 725	0.32	15 526	0.50	10 949
	DJFMAM	0.23	30 615	0.31	12 180	0.23	32 492	0.35	15 166	0.20	6819	0.37	4829
	JJASON	0.45	43 837	0.56	17 123	0.44	45 839	0.55	22 723	0.37	8194	0.55	5750
Central	All	0.39	39 942	0.47	18 584	0.36	40 824	0.51	21 084	0.30	8396	0.46	6256
	DJFMAM	0.27	15 892	0.31	7507	0.22	15 853	0.29	8130	0.23	3536	0.35	2549
	JJASON	0.45	23 217	0.55	10 708	0.44	23 979	0.57	12 506	0.33	4649	0.53	3551
Mountain	All	0.09	14 160	0.21	5007	0.07	15 597	0.13	6313	0.04	3455	0.06	2489
	DJFMAM	0.06	4788	0.00	1180	0.04	5258	−0.04	1463	−0.01	1385	−0.05	782
	JJASON	0.13	9178	0.30	3775	0.13	10 078	0.29	4793	0.12	1974	0.16	1659
Pacific	All	0.13	21 871	0.33	11 446	0.12	22 405	0.33	11 470	0.16	4639	0.27	3625
	DJFMAM	0.00	9110	0.08	4218	−0.03	9308	0.08	4265	0.06	2047	0.16	1509
	JJASON	0.24	12 310	0.44	7107	0.24	12 470	0.43	7011	0.27	2431	0.37	2025
Contiguous US	All	0.31	152 167	0.43	64 719	0.29	159 636	0.45	77 592	0.26	32 016	0.40	23 319
	DJFMAM	0.15	60 405	0.21	25 085	0.12	62 911	0.22	29 024	0.15	13 787	0.26	9669
	JJASON	0.40	88 542	0.52	38 713	0.39	92 366	0.52	47 033	0.34	17 248	0.48	12 985

Table 3. Two-year (2008–2009) correlation coefficients of hourly $PM_{2.5}$ observations and 40 km average CALIOP extinction (both uncorrected and dry mass) at various 100 m a.g.l. atmospheric layers.

CALIOP Extinction Layer	Uncorrected CALIOP Extinction		Dry Mass CALIOP Extinction	
	Eastern	Pacific	Eastern	Pacific
0–100 m	0.35	0.72	0.33	0.71
100–200 m	0.62	0.73	0.66	0.72
200–300 m	0.57	0.72	0.69	0.74
300–400 m	0.54	0.61	0.63	0.59
400–500 m	0.69	0.58	0.70	0.56

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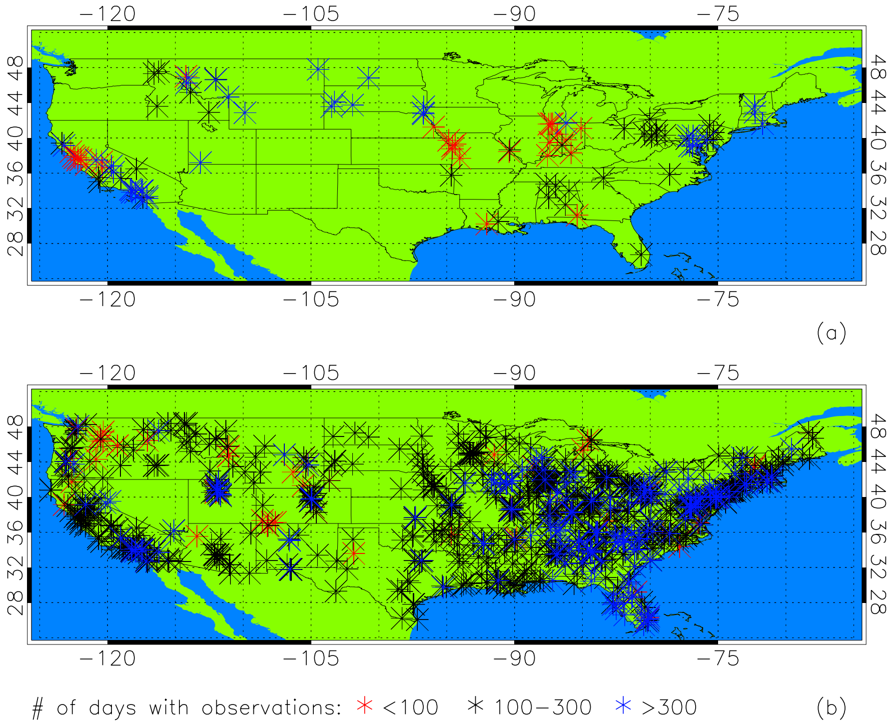

Fig. 1. For 2008–2009, US Environmental Protection Agency (EPA) sites with available $PM_{2.5}$ measurements at **(a)** hourly and **(b)** daily intervals, respectively. The sites are colored-coded based on number of days with observations, as red (fewer than 100), black (between 100 and 300), or blue (greater than 300).

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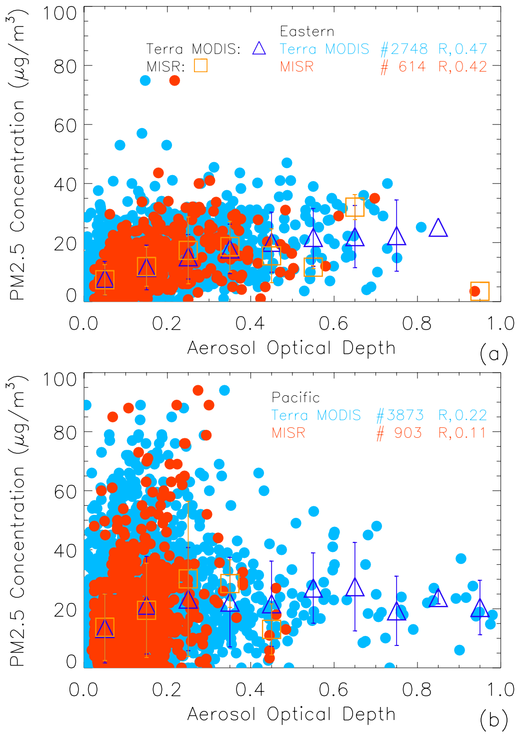

Fig. 2. Two-year (2008–2009) scatterplots of operational Terra MODIS (in light blue) and MISR (in red) AOD, averaged within 40 km of each respective $PM_{2.5}$-monitoring site, vs. hourly $PM_{2.5}$ concentrations for the **(a)** Eastern and **(b)** Pacific US time zones. Also plotted are averages of $PM_{2.5}$ for each 0.1 AOD bin, represented with triangles (in dark blue) for Terra MODIS and squares (in orange) for MISR. Error bars (±1 standard deviation) for the bin averages are also shown.

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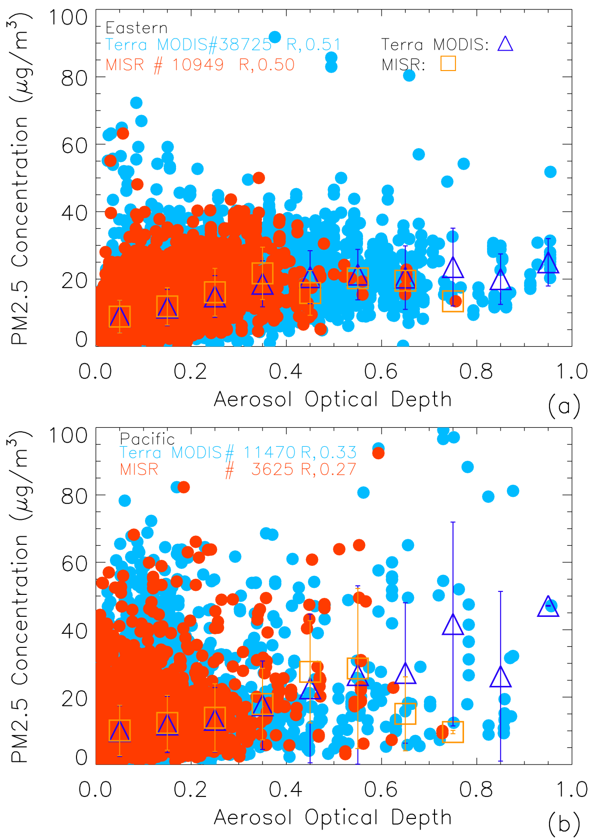

Fig. 3. Two-year (2008–2009) scatterplots of daily 1° × 1° DA Terra MODIS (in light blue) and daily 1° × 1° MISR (in red) AOD vs. daily $PM_{2.5}$ concentrations for the **(a)** Eastern and **(b)** Pacific US time zones. Averages of $PM_{2.5}$ are plotted for each 0.1 AOD bin, represented with triangles (in dark blue) for Terra MODIS and squares (in orange) for MISR. Error bars (±1 standard deviation) for the bin averages are also shown.

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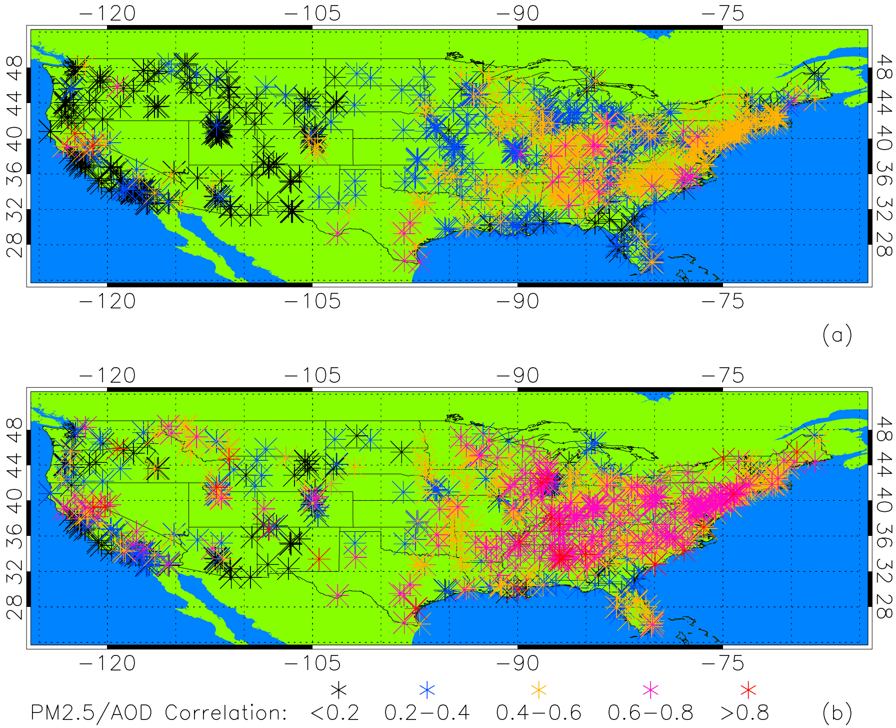

Fig. 4. For 2008–2009, those US Environmental Protection Agency (EPA) daily $PM_{2.5}$ sites used in this study. Sites are color-coded based on the correlation between daily $PM_{2.5}$ observations and daily 1° × 1° **(a)** operational and **(b)** DA Terra MODIS AOD.

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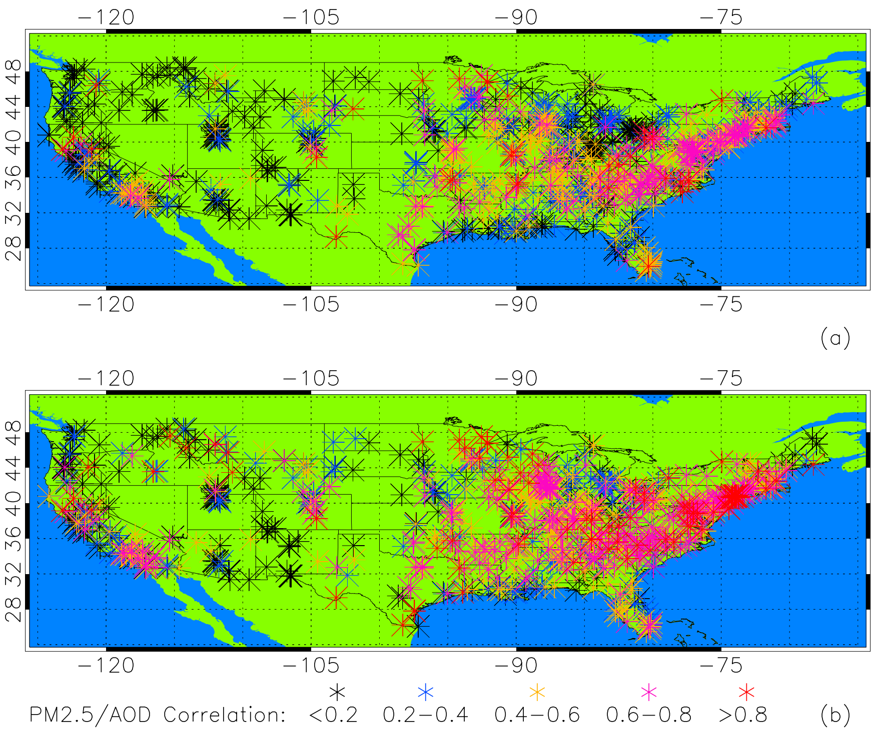

Fig. 5. For 2008–2009, US Environmental Protection Agency (EPA) daily $PM_{2.5}$ sites used in this study. Sites are color-coded based on the correlation between daily $PM_{2.5}$ observations and daily 1° × 1° **(a)** operational and **(b)** DA MISR AOD.

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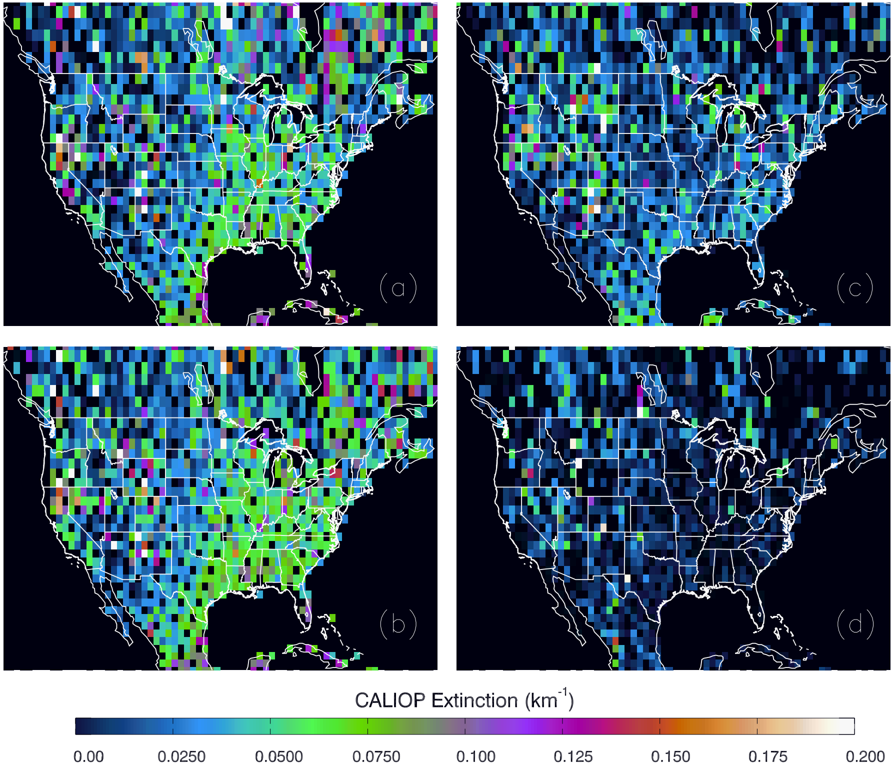

Fig. 6. Two-year (2008–2009) 1° × 1° average CALIOP 0.532 µm extinction, relative to the number of cloud free 5 km CALIOP profiles in each 1° × 1° bin, for atmospheric layers a.g.l. of **(a)** 0–500 m, **(b)** 500–1500 m, **(c)** 1500–2500 m, and **(d)** 2500–3500 m.

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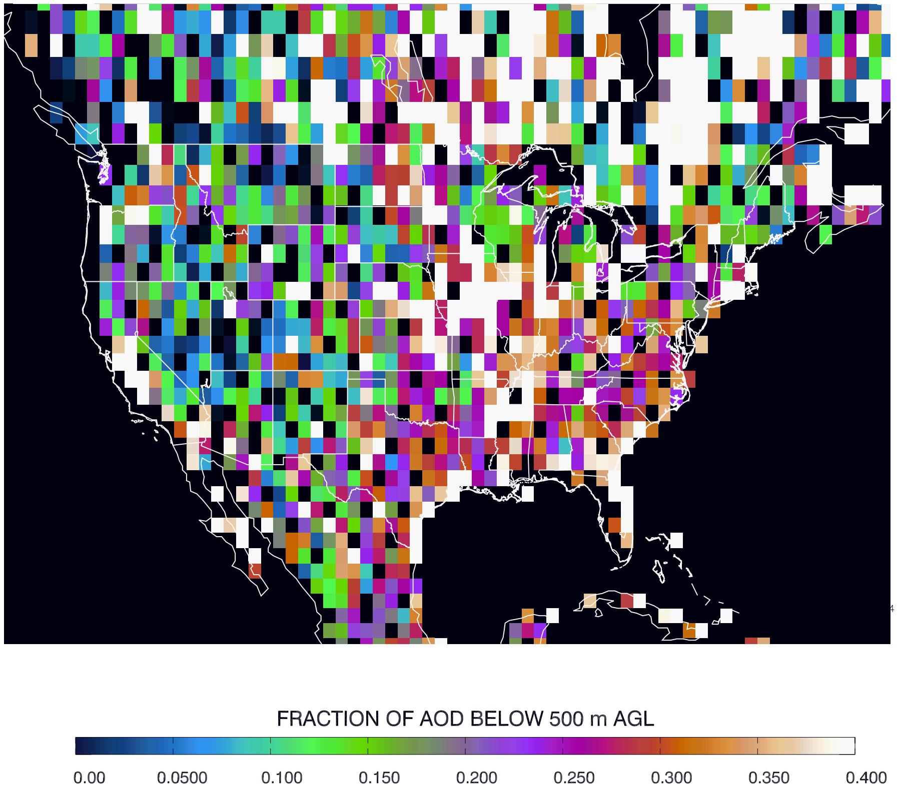

Fig. 7. Two-year (2008–2009) 1° × 1° average contribution percentage of 0 to 500 m a.g.l. integrated CALIOP extinction to total column AOD (at 0.532 µm), relative to the number of cloud free CALIOP profiles in each 1° × 1° bin, for the Continental United States.

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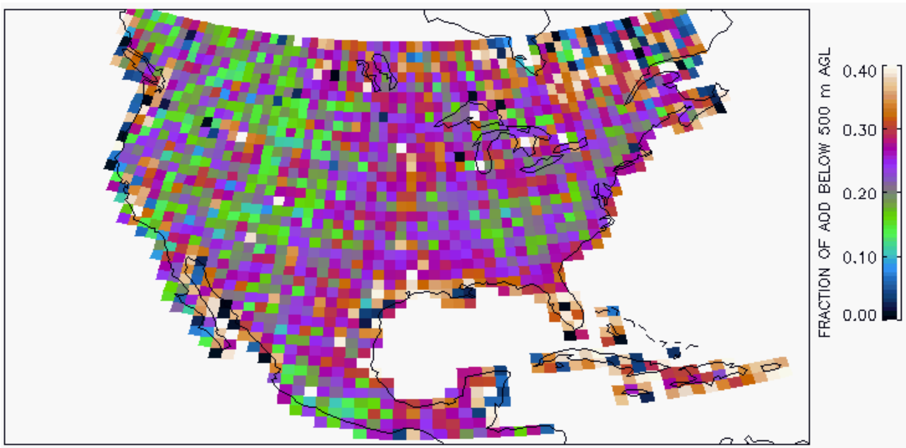

Fig. 8. From 2006–2011, fraction of CALIOP integrated 0.532 µm extinction below 500 m a.g.l. for the Continental United States.

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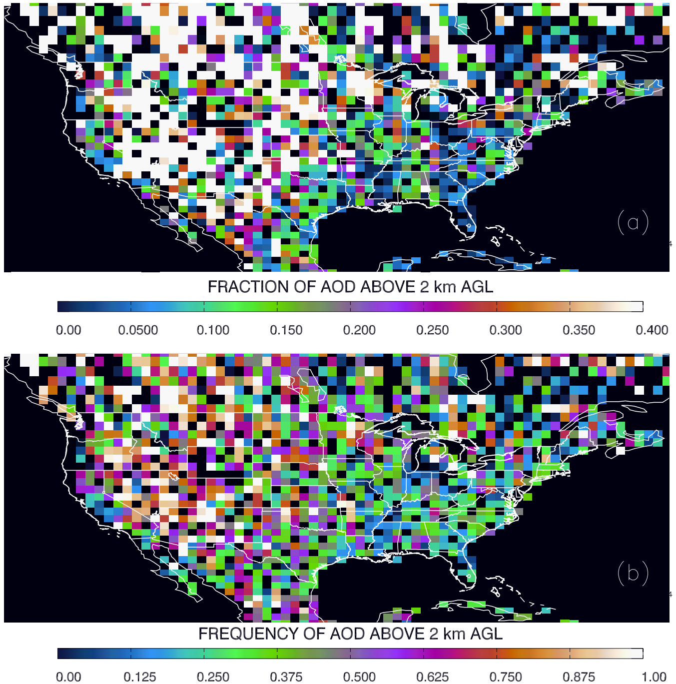

Fig. 9. Two-year (2008–2009) 1° × 1° average **(a)** contribution percentage of above 2 km a.g.l. CALIOP AOD to total column AOD (at 0.532 µm) and **(b)** frequency of occurrence of AOD above 2 km a.g.l., both relative to the number of cloud-free CALIOP profiles in each 1° × 1° bin, for the Continental United States.

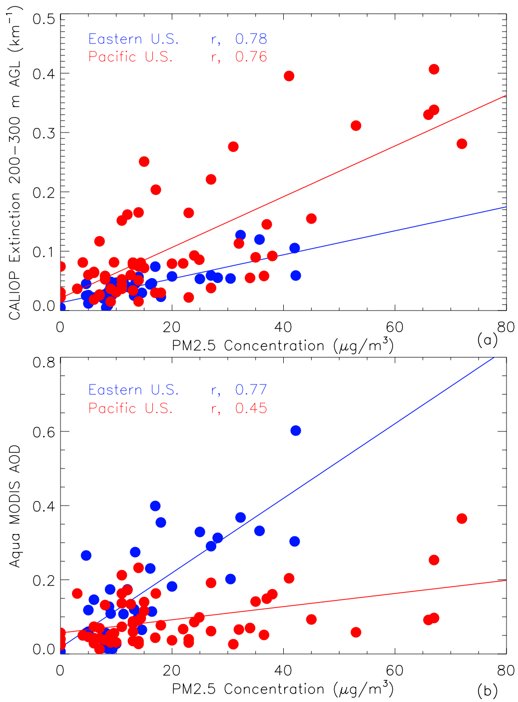

Fig. 10. For the Eastern (in blue) and Pacific (in red) US Time zones, two-year (2008–2009) scatterplots of hourly $PM_{2.5}$ concentrations vs. **(a)** cloud-free 5 km CALIOP dry mass 0.532 µm extinction at the 200–300 m a.g.l. layer, and **(b)** operational Aqua MODIS AOD, both averaged within 40 km and the hour of each respective $PM_{2.5}$ measurement.

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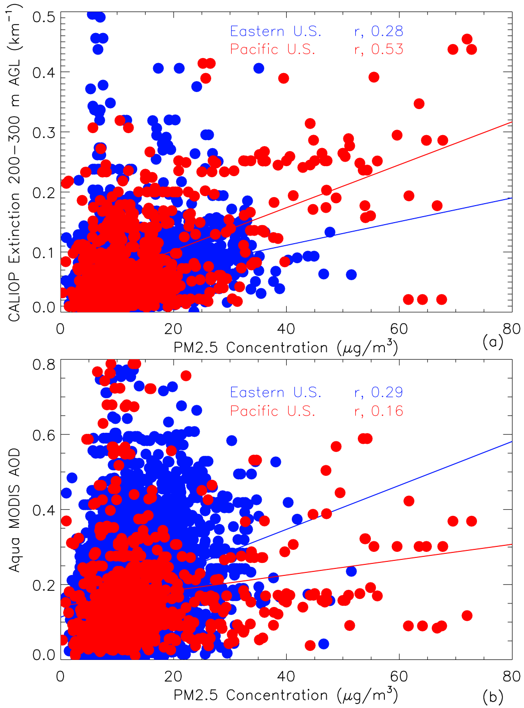

Fig. 11. For the Eastern (blue) and Pacific (red) US Time zones, two-year (2008–2009) scatter-plots of daily $PM_{2.5}$ concentrations vs. **(a)** cloud-free 5 km CALIOP dry mass 0.532 µm extinction at the 200–300 m a.g.l. layer (averaged within 100 km), and **(b)** operational Aqua MODIS AOD (averaged within 1°) and the day of each respective $PM_{2.5}$ measurement.

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