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A bulk-mass-modeling-based method for retrieving Particulate Matter Pollution using CALIOP observations

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Abstract. In this proof-of-concept paper, we apply a bulk-mass-modeling method using observations from the NASA Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP) instrument for retrieving particulate matter (PM) concentration over the contiguous United States (CONUS) over a 2-year period (2008-2009). Different from previous approaches that rely on empirical relationships between aerosol optical depth (AOD) and $PM_{2.5}$ (PM with particle sizes less than 2.5 μm), for the first time, we derive $PM_{2.5}$ concentrations, both at daytime and nighttime, from near surface CALIOP aerosol extinction retrievals using bulk mass extinction coefficients and model-based hygroscopicity. Preliminary results from this 2-year study conducted over the CONUS show a good agreement (r^2 ~ 0.48; mean bias of -3.3 μg m^{-3}) between the averaged nighttime CALIOP-derived $PM_{2.5}$ and ground-based $PM_{2.5}$ (with a lower r^2 of ~0.21 for daytime; mean bias of -0.4 μg m^{-3}), suggesting that PM concentrations can be obtained from active-based spaceborne observations with reasonable accuracy. Results from sensitivity studies suggest that accurate aerosol typing is needed for applying CALIOP measurements for $PM_{2.5}$ studies. Lastly, the e-folding correlation length for surface $PM_{2.5}$ is found to be around 600 km for the entire CONUS (~300 km for Western CONUS and ~700 km for Eastern CONUS), indicating that

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CALIOP observations, although sparse in spatial coverage, may still be applicable for $PM_{2.5}$ studies.

1 Introduction

During the last decade, an extensive number of studies have researched the feasibility of estimating $PM_{2.5}$ (particulate matter with particle sizes smaller than 2.5 μm) pollution with the use of passive-based satellite-derived aerosol optical depth (AOD; e.g., Liu et al., 2007; Hoff and Christopher, 2009; van Donkelaar et al., 2015). Monitoring of PM concentration from space observations is needed, as $PM_{2.5}$ pollution is one of the known causes of respiratory related diseases as well as other health related issues (e.g., Liu et al., 2005; Hoff and Christopher, 2009; Silva et al., 2013). Yet, ground-based $PM_{2.5}$ measurements are often inconsistent or have limited availability over much of the globe.

In some earlier studies, empirical relationships of $PM_{2.5}$ concentrations and AODs were developed and used for estimating $PM_{2.5}$ concentrations from passive sensor retrieved AODs (e.g., Wang and Christopher, 2003; Engel-Cox et al., 2004; Liu et al., 2005; Kumar et al., 2007; Hoff and Christopher, 2009). One of the limitations of this approach is that vertical distributions and thermodynamic state of aerosol particles vary with space and time. Especially for regions with elevated aerosol plumes, deep boundary layer entrainment zones, or strong nighttime inversions, column-integrated AODs are not a good approximation of surface $PM_{2.5}$ concentrations at specific points and times (e.g., Liu et al., 2004; Toth et al., 2014; Reid et al., 2017). Indeed, Kaku et al. (2018) recently showed that surface $PM_{2.5}$ had longer spatial correlation lengths than AOD, even in the "well behaved" southeastern United States where previous studies showed good performance (e.g., Wang and Christopher, 2003). To account for variability in aerosol vertical

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distribution, several studies have attempted the use of chemical transport models, or CTMs (e.g., van Donkelaar et al., 2015). Satellite data assimilation of AOD has become commonplace, vastly improving AOD analyses and short-term prediction (e.g., Zhang et al., 2014; Sessions et al., 2015). Yet, $PM_{2.5}$ simulations remain poor (e.g., Reid et al., 2016). Uncertainties in such studies are unavoidable due to uncertainties in CTM-based aerosol vertical distributions, and no nighttime AODs are currently available from passive-based satellite retrievals.

It is arguable that from a climatological/long-term average perspective, the use of AOD as a proxy for $PM_{2.5}$ concentrations nevertheless has certain qualitative skill (e.g., Toth et al., 2014; Reid et al., 2017) for the most significant events as well as due to the averaging process that suppresses sporadic aerosol events with highly variable vertical distributions. Still, as illustrated in Fig. 1, where 2-year (2008-2009) means of Moderate Resolution Imaging Spectroradiometer (MODIS) AOD are plotted against $PM_{2.5}$ concentrations throughout the contiguous United States (CONUS), although a linear relationship is plausibly shown, a low r^2 value of 0.09 is found. To construct Fig. 1, Aqua MODIS Collection 6 (C6) Optical_Depth_Land_And_Ocean data (0.55 μm), restricted to "Very Good" retrievals as reported by the Land_Ocean_Quality_Flag, are first collocated with daily surface $PM_{2.5}$ measurements in both space and time (i.e., within 40 km in distance and the same day), and then collocated daily pairs are averaged into 2-year means (for each $PM_{2.5}$ site). Figure 1 may be indicating that even from a long-term mean perspective, aerosol vertical distributions are not uniform across the CONUS, which is also confirmed by other studies (e.g., Toth et al., 2014). AOD retrievals themselves, with known uncertainties due to cloud contamination and assumptions in the retrieval process (e.g., Levy et al., 2013), may also introduce uncertainties to that task.

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77 On board the Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observations
78 (CALIPSO) satellite, the Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP) instrument
79 provides observations of aerosol and cloud vertical distributions at both day and night (Hunt et al.,
80 2009; Winker et al., 2010). Given that CALIOP provides aerosol extinction retrievals near the
81 ground, it is interesting and reasonable to raise the question: can near surface CALIPSO extinction
82 be used as a better physical quantity than AOD for estimating surface $PM_{2.5}$ concentrations? This
83 is because unlike AOD, which is a column-integrated value, near surface CALIPSO extinction is,
84 in theory, a more realistic representation of near surface aerosol properties. Yet, in comparing
85 with passive sensors such as MODIS, which has a swath width on the order of ~2000 km, CALIOP
86 is a nadir pointing instrument with a narrow swath of ~70 m and a repeat cycle of 16 days (Winker
87 et al., 2009). Thus, the spatial sampling of CALIOP is sparse on a daily basis and temporal
88 sampling or other conditional or contextual biases are unavoidable if CALIOP observations are
89 used to estimate daily $PM_{2.5}$ concentrations (Zhang and Reid, 2009; Colarco et al., 2014). Also,
90 there are known uncertainties in CALIPSO retrieved extinction values due to uncertainties in the
91 retrieval process, such as the lidar ratio (extinction-to-backscatter ratio), calibration, and the
92 "retrieval fill value" (RFV) issue (Young et al., 2013; Toth et al., 2018).

93 Even with these known issues, especially the sampling bias, it is still compelling to
94 investigate if near surface CALIOP extinction can be utilized to retrieve surface $PM_{2.5}$
95 concentrations with reasonable accuracy from a long-term (i.e., two-year) mean perspective.
96 CALIOP data have been successfully used in $PM_{2.5}$ studies in the past, but primarily for assisting
97 passive-based AOD/$PM_{2.5}$ analyses using aerosol vertical distribution as a constraint (e.g., Glantz
98 et al., 2009; van Donkelaar et al., 2010; Val Martin et al., 2013; Toth et al., 2014; Li et al., 2015;
99 Gong et al., 2017). However, the question remained as to the efficacy of the direct use of CALIOP

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retrievals. To demonstrate a concept, we developed a bulk mass scattering scheme for inferring PM concentrations from near surface aerosol extinction retrievals derived from CALIOP observations. The bulk method used here is based upon the well-established relationship between particle light scattering and $PM_{2.5}$ aerosol mass concentration (e.g., Charlson et al., 1968; Waggoner and Weiss, 1980; Liou, 2002; Chow et al., 2006), discussed further, with the relevant equations, in Sect. 2.

In this study, using two years (2008-2009) of CALIOP and United States (U.S.) Environmental Protection Agency (EPA) data over the CONUS, the following questions are addressed:

1. Can CALIOP extinction be used effectively for estimating $PM_{2.5}$ concentrations through a bulk mass scattering scheme from a 2-year mean perspective for both daytime and nighttime?
2. Can CALIOP extinction be used as a better parameter than AOD for estimating $PM_{2.5}$ concentrations from a 2-year mean perspective?
3. What are the sampling biases we can expect in CALIOP estimates of $PM_{2.5}$?
4. How do uncertainties in bulk properties compare to overall CALIOP-retrieved $PM_{2.5}$ uncertainty?

Details of the methods and datasets used are described in Sect. 2. Section 3 shows the preliminary results using two years of EPA $PM_{2.5}$ and CALIOP data, including an uncertainty analysis. The conclusions of this paper are provided in Sect. 4.

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2 Data and Methods

Since 1970, the U.S. EPA has monitored surface PM using a number of Federal Reference/Equivalent Methods (FRMs/FEMs), which employ gravimetric, tapered element oscillating microbalance (TEOM), and beta gauge instruments (Federal Register, 1997; Greenstone, 2002). Two years (2008-2009) of daily $PM_{2.5}$ Local Conditions (EPA code = 88101) data were acquired from the EPA Air Quality System for use in this investigation, consistent with our previous $PM_{2.5}$ study (Toth et al., 2014). We note that these data represent $PM_{2.5}$ concentrations over a 24-hour period and include two scenarios: one sample is taken during the 24-hour duration (i.e., filter-based measurement), or an average is computed from hourly samples within this time period (every hour may not have an available measurement, however).

CALIOP, flying aboard the CALIPSO platform within the A-Train satellite constellation, is a dual wavelength (0.532 and 1.064 μm) lidar that has collected profiles of atmospheric aerosol particles and clouds since summer 2006 (Winker et al., 2007). In this study, daytime and nighttime extinction coefficients retrieved at 0.532 μm from the Version 4.10 CALIOP Level 2 5 km aerosol profile (L2_05kmAPro) product were used. Using parameters provided in the L2_05kmAPro product, as well as the corresponding Level 2 5 km aerosol layer (L2_05kmALay) product, a robust quality-assurance (QA) procedure for the aerosol observations was implemented (Table 1). Further information on the QA metrics and screening protocol are discussed in detail in previous studies (Kittaka et al. 2011; Campbell et al. 2012; Toth et al. 2013; 2016). Once the QA procedure was applied, the aerosol profiles were linearly re-gridded from 60 m vertical resolution (above mean sea level [AMSL]) to 100 m segments (i.e., resampled to 100 m resolution) referenced to the local surface (above ground level [AGL]; Toth et al., 2014; 2016). The choice of 100 m was arbitrary, and the profiles were re-gridded in order to obtain an AGL-corrected dataset, as opposed

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146 to the AMSL-referenced profiles provided by the L2_05kmAPro product. Surface elevation and
147 relative humidity (RH) were taken from collocated model data included in the CALIPSO
148 L2_05kmAPro product (CALIPSO Data Products Catalog (Release 4.20); RH taken from the
149 Modern Era Retrospective-Analysis for Research, or MERRA-2 reanalysis product). To limit the
150 effects of signal attenuation and increase the chances of measuring aerosol presence near the
151 surface, the Atmospheric Volume Description parameter within the L2_05kmAPro dataset is used
152 to cloud-screen each aerosol profile as in Toth et al. (2018).

153 In this study, near surface PM mass concentration (C_m) is derived from near surface
154 CALIOP extinction based on a bulk formulation as in Equation 1 (e.g., Liou, 2002; Chow et al.,
155 2006):

156 $$\beta = C_m(a_{scat}f_{rh} + a_{abs}) \times 1000 \quad (1)$$

157 where β is CALIOP-derived near surface extinction in km^{-1}, C_m is the PM mass
158 concentration in $\mu g\ m^{-3}$, a_{scat} and a_{abs} are dry mass scattering and absorption efficiencies in $m^2\ g^{-1}$,
159 and f_{rh} represents the light scattering hygroscopicity, respectively. As a preliminary study, for the
160 purpose of demonstrating this concept, we assume the dominant aerosol type over the contiguous
161 U.S. (CONUS) is pollution aerosol (i.e., the most prevalent near-surface aerosol type reported in
162 the CALIOP products for the CONUS during 2008-2009 is polluted continental) with a_{scat} and a_{abs}
163 values of 3.40 and 0.37 $m^2\ g^{-1}$ (Hess et al., 1998; Lynch et al., 2016), respectively. These values
164 are similar to those reported in Malm and Hand (2007) and Kaku et al. (2018) but are interpolated
165 to 0.532 μm from values at 0.450 μm and 0.550 μm obtained from the Optical Properties of
166 Aerosols and Clouds (OPAC) model (Hess et al., 1998). Still, both a_{scat} and a_{abs} have regional and
167 species related dependencies. Also, only 2-year averages are used in this study, and we assume
168 that sporadic aerosol plumes are smoothed out in the averaging process, and that bulk aerosol

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properties are similar throughout the study region. We have further explored the impact of aerosol types to $PM_{2.5}$ retrievals in a later section. Furthermore, to aid in focusing this study on fine mode/anthropogenic aerosols, those aerosol extinction range bins classified as dust by the CALIOP typing algorithm were excluded from the analysis.

Also, surface PM concentrations are dry mass measurements. To account for the impact of humidity on a_{scat} (it is assumed that a_{abs} is not affected by moisture), we estimated the hygroscopic growth factor for pollution aerosol based on Hanel (1976), as shown in Equation 2:

$$f_{rh} = \left(\frac{1 - RH}{1 - RH_{ref}}\right)^{-\Gamma} \quad (2)$$

where f_{rh} is the hygroscopic growth factor, RH is the relative humidity, and RH_{ref} is the reference RH and is set to 30% in this study (Lynch et al., 2016). Γ is a unitless value (a fit parameter describing the amount of hygroscopic increase in scattering) and is assumed to be 0.63 (i.e., sulfate aerosol) in this study (Hanel, 1976; Chew et al., 2016; Lynch et al., 2016).

Lastly, the CALIOP-derived PM density is for all particle sizes. To convert from mass concentration of PM (C_m) to mass concentration of $PM_{2.5}$ ($C_{m2.5}$), which represents mass concentration for particle sizes smaller than 2.5 μm, we adopted the $PM_{2.5}$ to PM_{10} (PM with diameters less than 10 μm) ratio (ϕ) of 0.6 as measured during the Studies of Emissions and Atmospheric Composition, Clouds and Climate Coupling by Regional Surveys ($SEAC^4RS$) campaign over the US (Kaku et al., 2018). Again, the ratio of $PM_{2.5}$ to PM_{10} can also vary spatially, however we used a regional mean to demonstrate the concept. Analyses in a later section using two-years (2008-2009) of surface $PM_{2.5}$ to PM_{10} data suggest that 0.6 is a rather reasonable number to use for the CONUS for the study period. Here we assume that mass concentrations for particle sizes larger than 10 μm are negligible over the CONUS. Thus, we can rewrite Equation 1 as:

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$$C_{m2.5} = \frac{\beta \text{ x } \phi}{(a_{scat} \text{ x } f_{rh} + a_{abs}) \text{ x } 1000} \quad (3)$$

where $C_{m2.5}$ is the CALIOP-derived $PM_{2.5}$ concentration in units of μg m^{-3}.

3 Results and Discussion

3.1 Regional analysis

Figure 2a shows the mean $PM_{2.5}$ concentration using two years (2008-2009) of daily surface $PM_{2.5}$ data from the U.S. EPA ($PM_{2.5\_EPA}$), not collocated with CALIOP observations. A total of 1,091 stations (some operational throughout the entire period; others only partially) are included in the analysis and observations from those stations are further used in evaluating CALOP-derived $PM_{2.5}$ concentrations ($C_{m2.5}$), as later shown in Fig. 3. $PM_{2.5}$ concentrations of ~10 μg m^{-3} are found over the eastern CONUS. In comparison, much lower $PM_{2.5}$ concentrations of ~5 μg m^{-3} are exhibited for the interior CONUS, over states including Montana, Wyoming, North Dakota, South Dakota, Utah, Colorado, and Arizona. For the west coast of the CONUS, and especially over California, higher $PM_{2.5}$ concentrations are observed, with the maximum two-year mean near 20 μg m^{-3}. Note that the spatial distribution of surface $PM_{2.5}$ concentrations over the CONUS as shown in Fig. 2a is consistent with reported values from several studies (e.g., Hand et al., 2013; Van Donkelaar et al., 2015; Di et al., 2017).

Figure 3a shows the two-year averaged 1° x 1° (latitude/longitude) gridded daytime CALIOP aerosol extinction over the CONUS using CALIOP observations from 100-1000 m, referenced to the number of cloud-free L2_05kmAPro profiles in each 1 x 1° bin. The lowest 100 m of CALIOP extinction data are not used in the analysis due to the potential of surface return contamination (e.g., Toth et al., 2014), although this has been improved for the Version 4 CALIOP products but may still be present in some cases. Here the averaged extinction from 100-1000 m is

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used to represent near surface aerosol extinction. This selection of the 100-1000 m layer is somewhat arbitrary, even though it is estimated from the mean CALIOP-based aerosol vertical distribution over the CONUS (Toth et al., 2014), as surface layer heights may change seasonally and diurnally. Thus, a sensitivity study is provided in a later section to understand the impact of this aerosol layer selection to CALIOP-based $PM_{2.5}$ retrievals. As shown in Fig. 3a, higher mean near surface CALIOP extinction of 0.1 km^{-1} are found for the eastern CONUS and over California, while lower values of 0.025-0.05 km^{-1} found for the interior CONUS. Figure 3b shows a plot similar to Fig. 3a but using nighttime CALIOP observations only. Although similar spatial patterns are found during both day and night, the near surface extinction values are overall lower for nighttime than daytime, and nighttime data are less noisy than daytime. These findings are not surprising, as daytime CALIOP measurements are subject to contamination from background solar radiation (e.g., Omar et al., 2013).

To investigate any diurnal biases in the data, Figs. 3c and 3d show the derived $PM_{2.5}$ concentration using daytime and nighttime CALIOP data respectively, based on the method described in Section 2. Both Figures 3c and 3d suggest a higher $PM_{2.5}$ concentration of ~10-12.5 µg m^{-3} over the eastern CONUS, and a much lower $PM_{2.5}$ concentration of ~2.5-5 µg m^{-3} over the interior CONUS. High $PM_{2.5}$ values of 10-20 µg m^{-3} are also found over the west coast of the CONUS, particularly over California. The spatial distribution of $PM_{2.5}$ concentrations, as derived using near surface CALIOP data (Figs. 3c and 3d, as well as the combined daytime and nighttime perspective shown in Fig. 2c), is remarkably similar to the spatial distribution of $PM_{2.5}$ values as estimated based on ground-based observations (Fig. 2a). Still, day and night differences in $PM_{2.5}$ concentrations are also clearly visible, as higher $PM_{2.5}$ values are found, in general, during daytime, based on CALIOP observations. The high daytime $PM_{2.5}$ values, as shown in Fig. 3c, may

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represent stronger near surface convection and more frequent anthropogenic activities during daytime. However, they may also be partially contributed from solar radiation contamination. Another possibility is that the daytime mean extinction coefficients (from which the mean $PM_{2.5}$ estimates are derived) appear artifically larger than at night due to high daytime noise limiting the ability of CALIOP to detect fainter aerosol layers during daylight operations.

Figure 3e shows the inter-comparison between $PM_{2.5\_EPA}$ and $PM_{2.5\_CALIOP}$ concentrations. Note that only CALIOP and ground-based $PM_{2.5}$ data pairs, which are within 100 km of each other and have reported values for the same day (i.e., year, month, and day), are used to generate Fig. 3e. Still, although only spatially and temporally collocated data pairs are used, ground-based $PM_{2.5}$ data represent 24-hour averages, while CALIOP-derived $PM_{2.5}$ concentrations are instantaneous values over the daytime CALIOP overpass. To reduce this temporal bias, two years (2008-2009) of collocated CALIOP-derived and measured $PM_{2.5}$ concentrations are averaged and only the two-year averages are used in constructing Fig 3e. Also, to minimize the above-mentioned temporal sampling bias, ground stations with fewer than 100 collocated pairs are discarded. This leaves a total of 276 stations for constructing Fig. 3e.

As shown in Fig. 3e, an r^2 value of 0.21 (with a slope of 0.48) is found between CALIOP-derived and measured surface $PM_{2.5}$ concentrations, with a corresponding mean bias of -0.40 μg m^{-3} ($PM_{2.5\_CALIOP}$ - $PM_{2.5\_EPA}$). In comparison, Fig. 3f shows results similar to Fig. 3e, but for nighttime CALIOP data. A much higher r^2 value of 0.48 (with a slope of 0.67) is found between CALIOP-derived and measurement $PM_{2.5}$ values from 528 EPA stations, with a corresponding mean bias of -3.3 μgm^{-3} ($PM_{2.5\_CALIOP}$ - $PM_{2.5\_EPA}$). This may be related to the diurnal variability of $PM_{2.5}$ concentrations, as the daily mean EPA measurement might be closer to the CALIOP A.M. retrieval than to its P.M. counterpart. Still, data points are more scattered in Fig. 3e in comparison

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with Fig. 3f, which again indicates that daytime CALIOP data are noisier, possibly due to daytime solar contamination as well as other factors such as biases in relative humidity. Details of these biases are further explored in Section 3.2.

To supplement this analysis, a pairwise $PM_{2.5\_EPA}$ and $PM_{2.5\_CALIOP}$ (day and night CALIOP combined) analysis is presented in the spatial plots of Figs. 2b and 2d. Here, however, we lift the 100 collocated pairs requirement to increase data samples for better spatial representativeness. The spatial variability of $PM_{2.5}$ over the CONUS is consistent with the observed patterns of non-collocated data (i.e., Figs. 2a and 2c), but with generally higher values due to differences in sampling. Also, comparing Figs. 2b and 2d, $PM_{2.5\_EPA}$ spatial patterns match well with those of $PM_{2.5\_CALIOP}$, yet with larger values for $PM_{2.5\_EPA}$ (consistent with the biases discussed above). Lastly, a scatterplot of the pairwise analysis shown in Figs. 2b and 2d is provided in Fig. 4. An r^2 value of 0.40 is found between EPA and CALIOP-derived $PM_{2.5}$ concentrations from a combined daytime and nighttime CALIOP perspective. Overall, Figs. 2, 3, and 4 indicate that near surface CALIOP extinction data can be used to estimate surface $PM_{2.5}$ concentrations with reasonable accuracy.

3.2 Uncertainty analysis

In this section, uncertainties in the CALIOP derived, 2-year averaged $PM_{2.5}$ concentrations are explored as functions of aerosol vertical distribution, $PM_{2.5}$ to PM_{10} ratio, RH, aerosol type, and cloud presence above. Spatial sampling related biases as well as prognostic errors are also studied.

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283 **3.2.1 Prognostic errors in $C_{m2.5}$**

284 As a first step for the uncertainty analysis, we estimated the prognostic error of 2-year
285 averaged $PM_{2.5\_CALIOP}$. Figure 5 shows the root-mean-square error (RMSE) of CALIOP-based
286 $PM_{2.5}$ concentrations against those from EPA stations as a function of CALIOP-based $PM_{2.5}$ for
287 the 2008-2009 period over the CONUS. RMSEs were computed in intervals of 5 μg m^{-3} from 0
288 to 25 μg m^{-3}, with no computations greater than 25 μg m^{-3} performed due to very few data points
289 above this $PM_{2.5}$ concentration level. A mean combined daytime and nighttime minimum error of
290 ~4 μg m^{-3} is found, with generally larger RMSEs for nighttime below 15 μg m^{-3}, and larger RMSEs
291 for daytime above 15 μg m^{-3}. However, mean RMSEs (i.e., computed from the RMSEs shown in
292 Fig. 5) are similar for both datasets, ~4.5 μg m^{-3} for daytime and ~4.0 μg m^{-3} for nighttime. Also,
293 note that while the absolute error for daytime is largest at high $PM_{2.5}$ concentrations, relative errors
294 are similar (e.g., 3 μg m^{-3}/10 μgm^{-3} or 30% for the 5-10 μg m^{-3} bin, versus 7 μg m^{-3}/25 μg m^{-3} or
295 28% for the 20-25 μg m^{-3} bin). For context, the number of samples per bin are also plotted (as X
296 symbols) in Fig. 5. Data sample sizes are smallest (largest) for the lowest/highest range (mid-
297 range) $PM_{2.5}$ bins.
298
299 **3.2.2 Surface layer height sensitivity study**
300 A sensitivity study was conducted for which $PM_{2.5}$ was derived from near-surface CALIOP
301 aerosol extinction by varying the height of the surface layer in increments of 100 m from the
302 ground to 1000 m. Note that the surface layer (0-100 m) is included for this sensitivity study only.
303 The statistical results of this analysis, for both daytime and nighttime conditions, are shown in
304 Table 2. Four statistical parameters were computed, consisting of r^2, slope, mean bias (CALIOP
305 – EPA) of $PM_{2.5}$, and percent error change in derived $PM_{2.5}$, defined as: ((mean_new_$PM_{2.5}$ –

306 mean_original_$PM_{2.5}$)/mean_original_$PM_{2.5}$)*100. For context, the bottom row of Table 2 shows
307 the results from the original analysis. In terms of r^2 and slope, optimal values peak at different
308 surface layer heights between daytime and nighttime. For example, for daytime, the largest
309 correlations are found for the 0-600 m and 0-700 m layers, while for nighttime these are found for
310 the 0-300 m and 0-400 m layers. However, the 0-300 m layer (100-1000 m layer) exhibits the
311 lowest mean bias for the daytime (nighttime) analysis. Overall, marginal changes are found for
312 varying the height of the surface layer. Yet the largest mean bias is found for the 0-100 m layer,
313 indicating the need for excluding the 0-100 m layer in the analysis.
314
315 **3.2.3 RH sensitivity study**
316 Profiles of RH were taken from the MERRA-2 reanalysis product, as these collocated data
317 are provided in the CALIPSO L2_05kmAPro product. However, biases may exist in this RH
318 dataset. Thus, we examined the impact of varying the RH values by +/- 10% on the CALIOP-
319 derived $PM_{2.5}$ concentrations. For both daytime and nighttime analyses, no significant differences
320 in the r^2 and slope values were found. However, a +15% (-15%) change in the mean derived $PM_{2.5}$
321 values was found by decreasing (increasing) the RH values by 10%.
322
323 **3.2.4 $PM_{2.5}$ to PM_{10} ratio sensitivity study**
324 Another source of uncertainty in this study is the $PM_{2.5}/PM_{10}$ ratio. Using surface-based
325 $PM_{2.5}$ and PM_{10} data from those EPA stations over the CONUS for 2008-2009 with concurrent
326 $PM_{2.5}$ and PM_{10} daily data available (i.e., 409 stations), we computed the mean $PM_{2.5}/PM_{10}$ ratio,
327 and its corresponding standard deviation. The mean ratio was 0.56 with a standard deviation of
328 0.32. It is interesting to note that the mean $PM_{2.5}/PM_{10}$ ratio estimated from two years of surface

329 observations over the CONUS is close to 0.6 (the number used in this study), as reported by Kaku
330 et al. (2018). We also tested the sensitivity of the derived $PM_{2.5}$ concentrations as a function of
331 $PM_{2.5}/PM_{10}$ ratio for two scenarios: ±1 standard deviation of the mean (Table 3). In general, a ±50
332 % to 60 % change is found with the variation of the $PM_{2.5}/PM_{10}$ ratio at the range of ±1 standard
333 deviation of the mean. As suggested from Table 3, the optimal slope is found using a ratio of +1
334 standard deviation of the mean for both daytime and nighttime. The lowest mean daytime bias is
335 found for a ratio of 0.6, and for nighttime the lowest mean bias occurs using a ratio of 0.88.
336
337 **3.2.5 Sampling-related biases**
338 As mentioned in the introduction section, a sampling bias, due to the very small footprint
339 size and ~16 day repeat cycle of CALIOP, can exist when using CALIOP observations for $PM_{2.5}$
340 estimates (Zhang and Reid, 2009). This sampling-induced bias is investigated from a 2-year mean
341 perspective by comparing histograms of $PM_{2.5\_EPA}$ and $C_{m2.5}$ concentrations as shown in Fig. 6. To
342 generate Fig. 6, all available daily EPA $PM_{2.5}$ are used to represent the "true" 2-year mean spectrum
343 of $PM_{2.5}$ concentrations over the EPA sites. The aerosol extinction data spatially collocated to the
344 EPA sites (Sect. 3.1), but not temporally collocated, are used for estimating the 2-year mean
345 spectrum of $PM_{2.5}$ concentrations as derived from CALIOP observations. To be consistent with
346 the previous analysis, only cloud-free CALIOP profiles are considered. The $PM_{2.5\_EPA}$
347 concentrations peak at ~9 μg m^{-3} (standard deviation of ~3 μg m^{-3}), and CALIOP-derived $PM_{2.5}$
348 peaks at ~9 μg m^{-3} (daytime; standard deviation of ~4 μg m^{-3}) and ~5 μg m^{-3} (nighttime; standard
349 deviation of ~2 μg m^{-3}). The distribution shifts towards smaller concentrations for CALIOP, more
350 so for nighttime than daytime (possibly due to CALIOP daytime versus nighttime detection
351 differences).

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Still, Fig. 6 may reflect the diurnal difference in $PM_{2.5}$ concentrations as well as the retrieval bias in $C_{m2.5}$ values. Thus, we have re-performed the exercise shown in Fig. 6 using spatially and temporally collocated $PM_{2.5\_EPA}$ and $C_{m2.5}$ data as shown in Fig. 7. To construct Fig. 7, $PM_{2.5\_EPA}$ and $C_{m2.5}$ data are collocated following the steps mentioned in Sect. 3.1, with CALIOP and EPA $PM_{2.5}$ representing 2-year mean values for each EPA station. Again, only cloud-free CALIOP profiles are considered for this analysis. As shown in Fig. 7a, the $PM_{2.5\_EPA}$ concentrations peak at ~7 μg m^{-3} (standard deviation of ~4 μg m^{-3}), and daytime $C_{m2.5}$ peaks at ~6 μg m^{-3} (standard deviation of ~4 μg m^{-3}). In comparison, with the use of collocated nighttime $C_{m2.5}$ and $PM_{2.5\_EPA}$ data as shown in Fig. 7b, the peak $PM_{2.5\_EPA}$ value is about 2 μg m^{-3} higher than the peak $C_{m2.5}$ value (with similar standard deviations as found in the analyses of Fig. 7a). Considering both Figs. 6 and 7, it is likely that the temporal sampling bias seen in Fig. 6 is at least in part due to retrieval bias as well as the difference in $PM_{2.5}$ concentrations during daytime and nighttime.

3.2.6 CALIOP AOD analysis

Most past studies focused on the use of column AODs as proxies for surface $PM_{2.5}$ (e.g., Liu et al., 2005; Hoff and Christopher, 2009; van Donkelaar et al., 2015). Therefore, it is interesting to investigate whether near surface CALIOP extinction values can be used as a better physical quantity to estimate surface $PM_{2.5}$ in comparing with column-integrated CALIOP AOD. To achieve this goal, we have compared CALIOP column AOD and $PM_{2.5}$ from EPA stations, as shown in Fig. 8. Similar to the scatterplots of Fig. 4, each point represents a two-year mean for each EPA site, and was created from a dataset following the same spatial/temporal collocation as described above. As shown in Fig. 9, r^2 values of 0.04 and 0.13 are found using CALIOP daytime

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375 and nighttime AOD data, respectively, similar to the MODIS-based analysis shown in Fig. 1. This
376 is expected, as elevated aerosol layers will negatively impact the relationship between surface
377 $PM_{2.5}$ and column AOD. The derivation of surface $PM_{2.5}$ from near surface CALIOP extinction,
378 as demonstrated from this study however, provides a much better spatial matching between the
379 quantities being compared, with potential error terms that can be well quantified and minimized in
380 later studies.
381
382 **3.2.7 Cloud flag sensitivity study**
383 For most of this paper, a strict cloud screening process is implemented, during which no
384 clouds are allowed in the entire CALIOP profile. However, in contrast to passive sensor
385 capabilities (e.g., MODIS), near-surface aerosol extinction coefficients can be readily retrieved
386 from CALIOP profiles even when there are transparent cloud layers above. Therefore, we
387 conducted an additional analysis for which no cloud flag was set (i.e., all-sky conditions). Results
388 are shown in scatterplot form in Fig. 9, in a similar manner as Figs. 3e and f, with an additional 97
389 (156) points for the daytime (nighttime) analyses. Comparing the all-sky results with those of
390 Figs. 3e, and f (cloud-free conditions), the r^2 values are similar. This is also true in terms of mean
391 bias, with similar values of 0.70 (-2.68) μg m^{-3} found for daytime (nighttime) for all-sky scenarios.
392 This indicates that our method performs reasonably well from an all-sky perspective. However,
393 we note that restricting the analysis to solely those cases that are cloudy (not shown), the method
394 does not perform as well. For example, the r^2 values decrease by 71% (90%) and the slope values
395 decrease by 21% (75%) for the daytime (nighttime) analyses, compared to the cloud-free results
396 (Figs. 3e and f). This is expected, as any errors made in estimating the optical depths of the

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397 overlying clouds will propagate (as biases) into the extinction retrievals for the underlying
398 aerosols.
399
400 **3.2.8 Aerosol type analysis**
401 Also, for this study, we assume that the primary aerosol type over the CONUS is pollution
402 (i.e., sulfate & organic) aerosol, which is generally composed of smaller (fine mode) particles that
403 tend to exhibit mass extinction efficiencies ~4 $m^2 g^{-1}$. However, even after implementing our dust-
404 free restriction, the study region can also be contaminated with non-pollution aerosols, which can
405 have a larger particle size and exhibit lower mass extinction efficiencies (e.g., Hess et al., 1998;
406 Malm and Hand, 2007; Lynch et al., 2016). The use of $PM_{2.5}$ versus PM_{10} somewhat mitigates
407 this size dependency, but nevertheless coarse mode dust or sea salt can dominate $PM_{2.5}$ mass values
408 (e.g., Atwood et al., 2013).
409 Thus, in this section, the impact of aerosol types to the derived $PM_{2.5}$ concentrations was
410 explored by varying the mass scattering and absorption efficiencies and gamma values associated
411 with each aerosol type. The three aerosol types chosen for this sensitivity study were dust, sea
412 salt, and smoke, based upon Lynch et al. (2016). The mass scattering and absorption values for
413 dust and sea salt were interpolated to 0.532 µm from values at 0.450 µm and 0.550 µm from OPAC
414 (as was done for the sulfate case; Hess et al., 1998). For smoke, these values were interpolated to
415 0.532 µm from values at 0.440 µm and 0.670 µm as provided by Reid et al. (2005) for smoke cases
416 over the US and Canada. The gamma values were taken from Lynch et al. (2016) and the
417 references within. These values, as well as the results from this sensitivity study, are shown in
418 Table 4. If we assume all aerosols within the study region are smoke aerosols, no major changes
419 in the retrieved CALIOP $PM_{2.5}$ values are found. However, significant uncertainties on the order

of ~200% (~800%) are found if sea salt (dust) aerosol mass scattering/absorption efficiencies and gamma values are used instead. Clearly, this study suggests that accurate aerosol typing is necessary for future applications of CALIOP observations for surface $PM_{2.5}$ estimations.

3.2.9 E-folding correlation length for $PM_{2.5}$ concentrations over the CONUS

As a last study, we also estimated the spatial e-folding correlation length for $PM_{2.5}$ concentrations over the CONUS. This provides us an estimation of the correlation between a CALIOP-derived and actual $PM_{2.5}$ concentration for a given location as a function of distance between the CALIOP observation and the given location. To accomplish this, for 2008-2009 over the CONUS, the distances and correlations (of $PM_{2.5}$ concentration) were computed for any two EPA stations with over 50 days of daily data for the two-year period. Results are shown in Fig. 10 as a scatterplot, with individual points in gray and the black curve representing the exponential fit to the data. A decrease in $PM_{2.5}$ correlation with distance between EPA stations is found, and the e-folding length in correlation (e.g., correlation reduced to 1/e, or 0.37) is ~600 km (from an AOD standpoint, this value is 40-400 km, as suggested by Anderson et al., 2003).

Also included in Fig. 10 are results from a corresponding regional analysis, with the red and blue lines showing bin averages (10 km) for the Western and Eastern CONUS, respectively (regions partitioned by the -97° longitude line). The e-folding length is ~300 km (~700 km) for the Western (Eastern) CONUS, indicating a much shorter correlation length for pollution over the Western CONUS, possibly due to a more complex terrain such as mountains. Overall these $PM_{2.5}$ e-folding lengths suggest that CALIOP-derived $PM_{2.5}$ concentrations could still have some representative skill within a few hundred kilometers of a given location.

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4 Conclusions

In this paper, we have demonstrated a new bulk-mass-modeling method for retrieving surface particulate matter (PM) with particle sizes smaller than 2.5 μm ($PM_{2.5}$) using observations acquired by the NASA Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP) instrument from 2008-2009. For the purposes of demonstrating this concept, only regionally-averaged parameters, such as mass scattering and absorption coefficients, and $PM_{2.5}$ to PM_{10} (PM with particle sizes smaller than 10 μm) conversion ratio, are used. Also, we assume the dominant type of aerosols over the study region is pollution aerosols (supported by the occurrence frequencies of aerosol types determined by the CALIOP algorithms), and exclude aerosol extinction range bins classified as dust from the analysis. Even with the highly-averaged parameters, the results from this paper are rather promising and demonstrate a potential for monitoring PM pollution using active-based lidar observations. Specifically, the primary results of this study are as follows:

1. CALIOP-derived $PM_{2.5}$ concentrations of ~10-12.5 μg m^{-3} are found over the eastern contiguous United States (CONUS), with lower values of ~2.5-5 μg m^{-3} over the central CONUS. $PM_{2.5}$ values of ~10-20 μg m^{-3} are found over the west coast of the CONUS, primarily California. The spatial distribution of 2-year mean $PM_{2.5}$ concentrations derived from near surface CALIOP aerosol data compares well to the spatial distribution of *in situ* $PM_{2.5}$ measurements collected at the ground-based stations of the U.S. Environmental Protection Agency (EPA). The use of nighttime CALIOP extinction to derive $PM_{2.5}$ results in a higher correlation (r^2 = 0.48; mean bias = -3.3 μgm^{-3}) with EPA $PM_{2.5}$ than daytime CALIOP extinction data (r^2 = 0.21; mean bias = -0.40 μgm^{-3}).
2. Correlations between CALIOP aerosol optical depth (AOD) and EPA $PM_{2.5}$ are much lower (r^2 values of 0.04 and 0.13, for daytime and nighttime CALIOP AOD data,

466 respectively) than those obtained from derived $PM_{2.5}$ using near-surface CALIOP aerosol
467 extinction. A similar correlation is also found between Moderate Resolution Imaging
468 Spectroradiometer (MODIS) AOD and EPA $PM_{2.5}$ from two-year (2008-2009) means.
469 This suggests that CALIOP extinction may be used as a better parameter for estimating
470 $PM_{2.5}$ concentrations from a 2-year mean perspective. Also, the algorithm proposed in this
471 study is essentially a semi-physical-based method, and thus the retrieval process can be
472 improved, upon a careful study of the physical parameters used in the process.
473 3. Spatial and temporal sampling biases, as well as a retrieval bias, are found. Also, several
474 sensitivity studies were conducted, including surface layer height, cloud flag, $PM_{2.5}/PM_{10}$
475 ratio, relative humidity, and aerosol type. The sensitivity studies highlight the need for
476 accurate aerosol typing for estimating $PM_{2.5}$ concentrations using CALIOP observations.
477 4. Using surface-based $PM_{2.5}$ at EPA stations alone, the e-folding correlation length for $PM_{2.5}$
478 concentrations was found to be about 600 km for the CONUS. A regional analysis yielded
479 values of ~300 km and ~700 km for the Western and Eastern CONUS, respectively. Thus,
480 while limited in spatial sampling, measurements from CALIOP may still be used for
481 estimating $PM_{2.5}$ concentrations over the CONUS.

482 As noted earlier, CALIOP observations are still rather sparse, and concerns related to
483 reported CALIOP aerosol extinction values also exist, such as solar and surface contamination and
484 the "retrieval fill value" issue (e.g., Toth et al., 2018). Yet, the future High Spectral Resolution
485 Lidar (HSRL) instrument on board the Earth Clouds, Aerosol, and Radiation Explorer
486 (EarthCARE) satellite (Illingworth et al., 2015), as well as forthcoming space-based lidar missions
487 in response to the 2017 Decadal Survey, offer opportunities to further explore aerosol extinction -

based PM concentrations. Ultimately the results from this study show that the combined use of several lidar instruments for monitoring regional and global PM pollution is potentially achievable.

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754 **Figure and Table Captions**
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756 Figure 1. For 2008-2009, scatterplot of mean $PM_{2.5}$ concentration from ground-based U.S. EPA
757 stations and mean column AOD (550 nm) from collocated Collection 6 (C6) Aqua MODIS
758 observations.

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760 Figure 2. For 2008-2009 over the CONUS, (a) mean $PM_{2.5}$ concentration (µg m^{-3}) for those U.S.
761 EPA stations with reported daily measurements, and (c) 1° x 1° average CALIOP-derived $PM_{2.5}$
762 concentrations for the 100–1000 m AGL atmospheric layer, using Equation 3, for combined
763 daytime and nighttime conditions. Also shown are the pairwise $PM_{2.5}$ concentrations from (b)
764 EPA daily measurements and (d) those derived from CALIOP (day and night combined), both
765 averaged for each EPA station for the 2008-2009 period. For all four plots, values greater than 20
766 µgm^{-3} are colored red.

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768 Figure 3. For 2008-2009 over the CONUS, 1° x 1° average CALIOP extinction, relative to the
769 number of cloud-free 5 km CALIOP profiles in each 1° x 1° bin, for the 100 – 1000 m AGL
770 atmospheric layer, for (a) daytime and (b) nighttime measurements. Also shown are the
771 corresponding CALIOP-derived $PM_{2.5}$ concentrations, using Equation 3 for (c) daytime and (d)
772 nighttime conditions. Values greater than 0.2 km^{-1} and 20 µg m^{-3} for (a, b) and (c, d), respectively,
773 are colored red. Scatterplots of mean $PM_{2.5}$ concentration from ground-based U.S. EPA stations
774 and those derived from collocated near-surface CALIOP observations are shown in the bottom
775 row, using (e) daytime and (f) nighttime CALIOP data.

776 Figure 4. Scatterplot of mean $PM_{2.5}$ concentration from ground-based U.S. EPA stations and those
777 derived from collocated near-surface CALIOP observations using combined daytime and
778 nighttime CALIOP data.
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780 Figure 5. Root-mean-square errors of CALIOP-derived $PM_{2.5}$ against EPA $PM_{2.5}$ as a function of
781 CALIOP-derived $PM_{2.5}$ (filled circles), and corresponding number of data samples per bin (X
782 symbols), using both daytime (in red) and nighttime (in blue) CALIOP observations.
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784 Figure 6. Two-year (2008-2009) histograms of mean $PM_{2.5}$ concentrations from the U.S. EPA (in
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789 Figure 7. Two-year (2008-2009) histograms of mean $PM_{2.5}$ concentrations from the U.S. EPA and
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791 (b) nighttime CALIOP data.
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798 stations and those derived from collocated all-sky (including cloud-free and cloudy profiles) near-
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801 Figure 10. For 2008-2009 over the CONUS, scatterplot of distance (km) between any two U.S.
802 EPA stations and the corresponding spatial correlation of $PM_{2.5}$ concentration between each pair
803 of stations. The black curve represents the exponential fit to the data for the entire CONUS, and
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821 derivation of $PM_{2.5}$, including R^2, slope, mean bias (CALIOP - EPA) of $PM_{2.5}$ in µg m^{-3}, and
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844 **Figures**

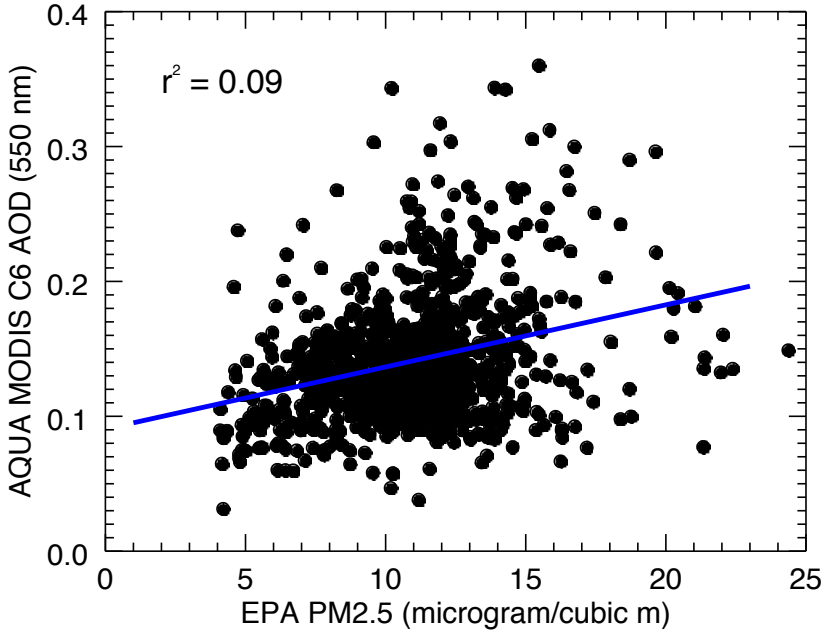

Figure 1. For 2008-2009, scatterplot of mean $PM_{2.5}$ concentration from ground-based U.S. EPA stations and mean column AOD (550 nm) from collocated Collection 6 (C6) Aqua MODIS observations.

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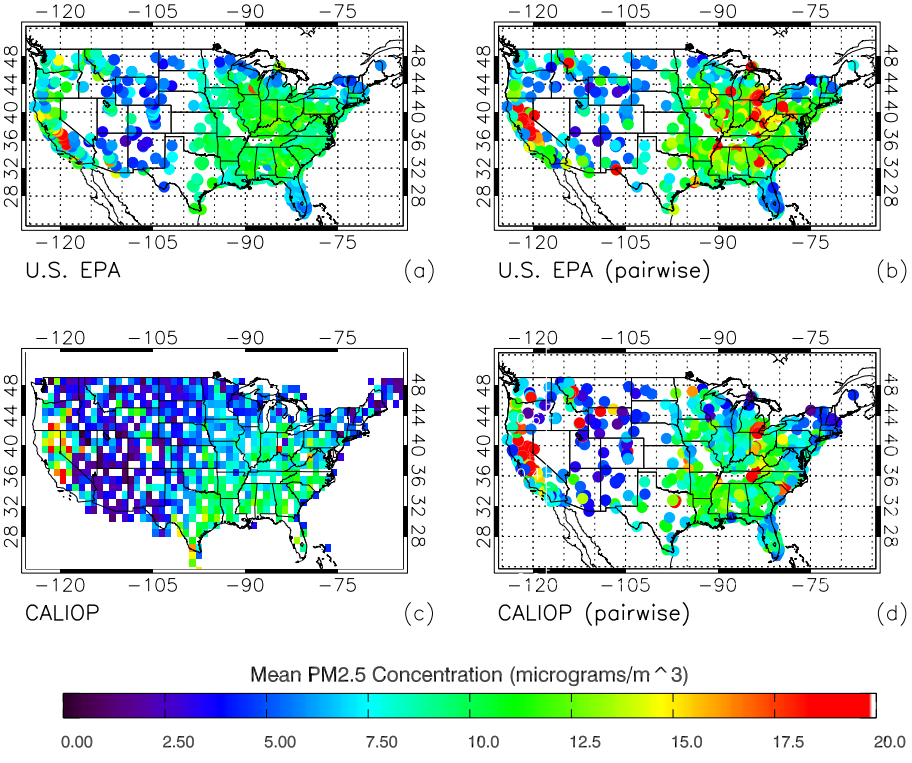

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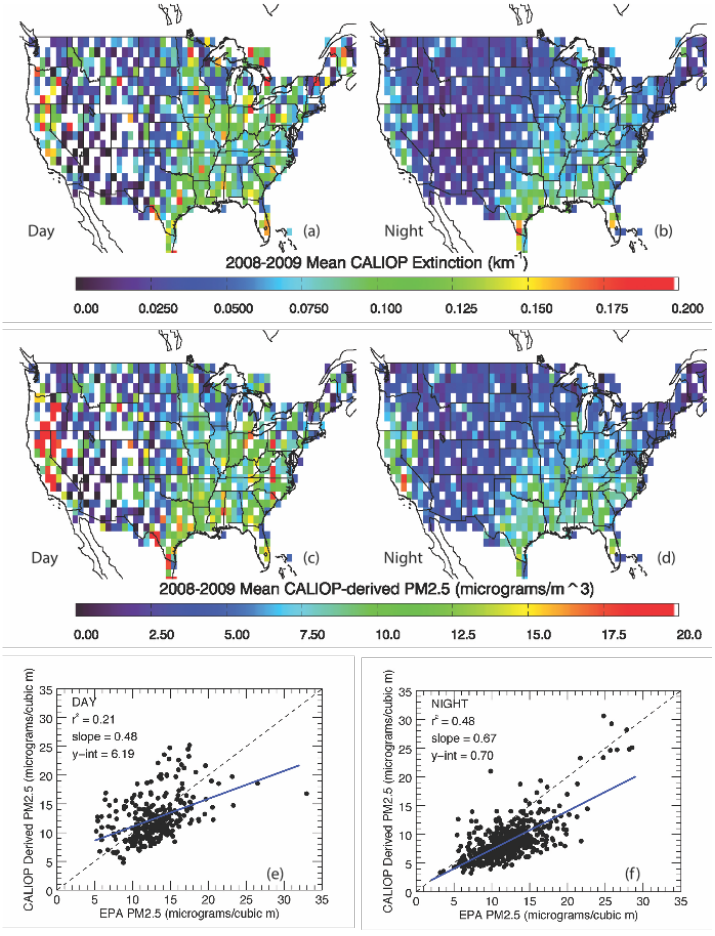

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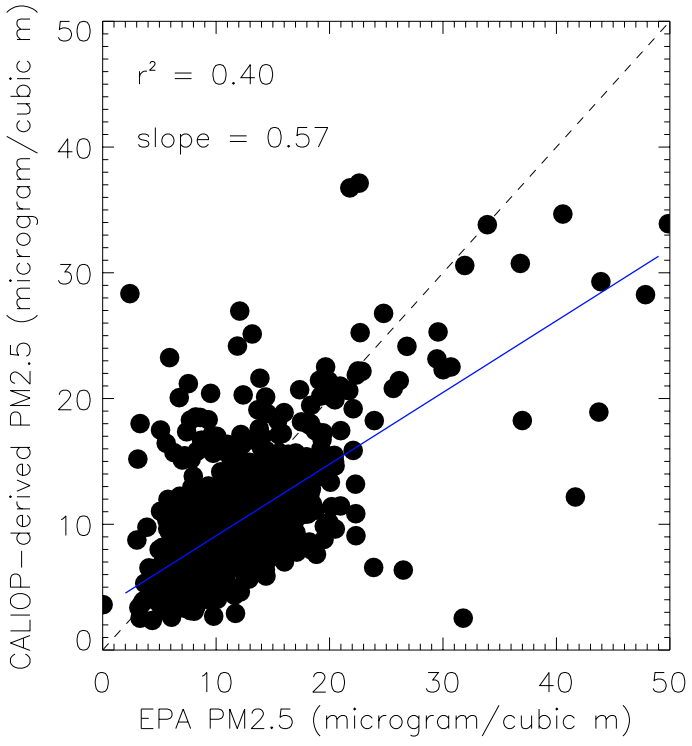

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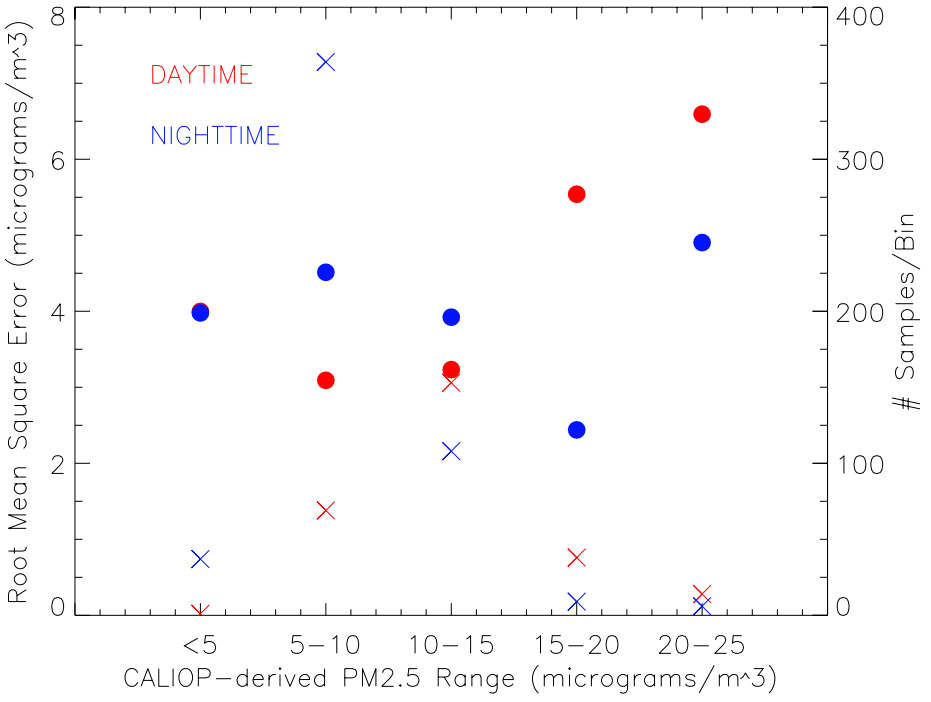

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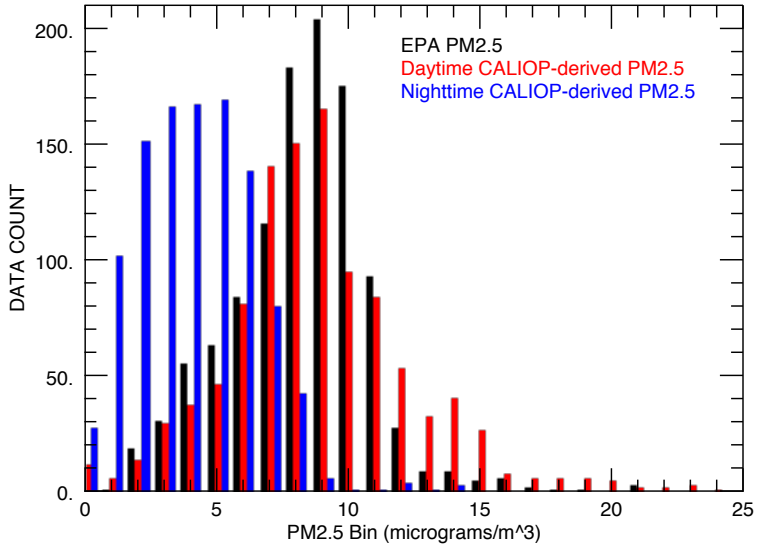

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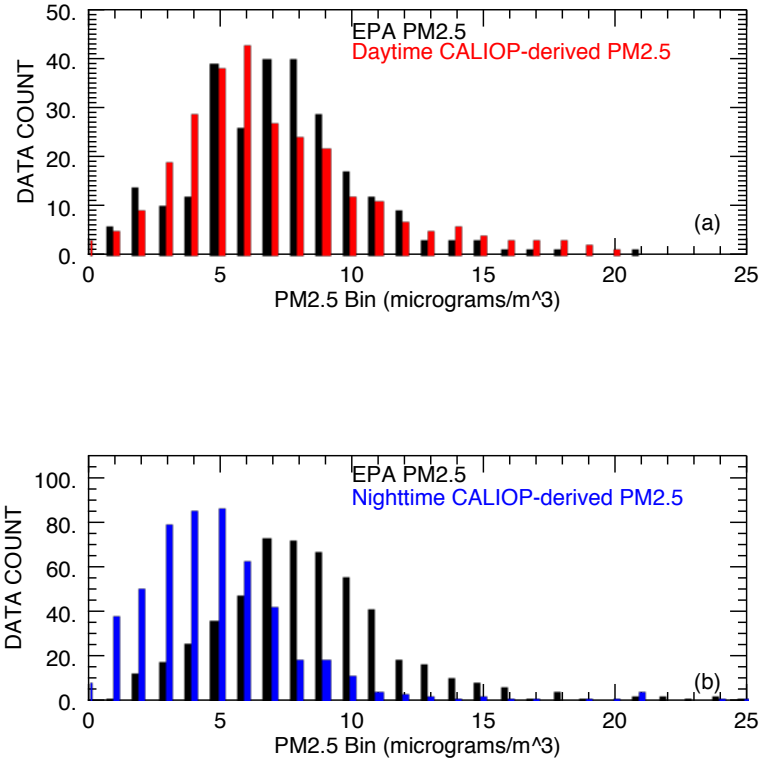

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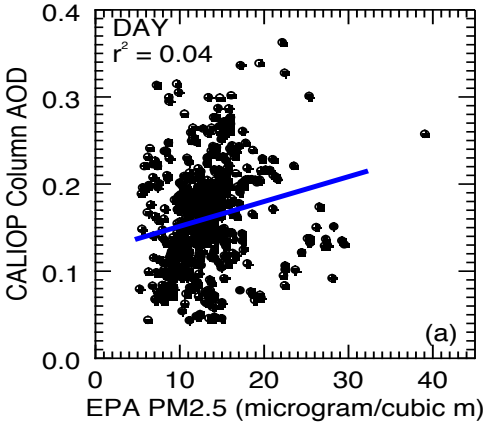

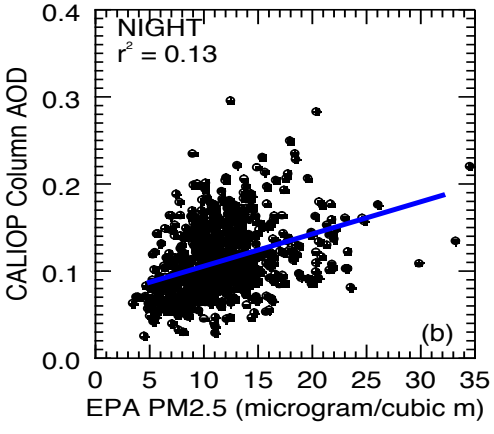

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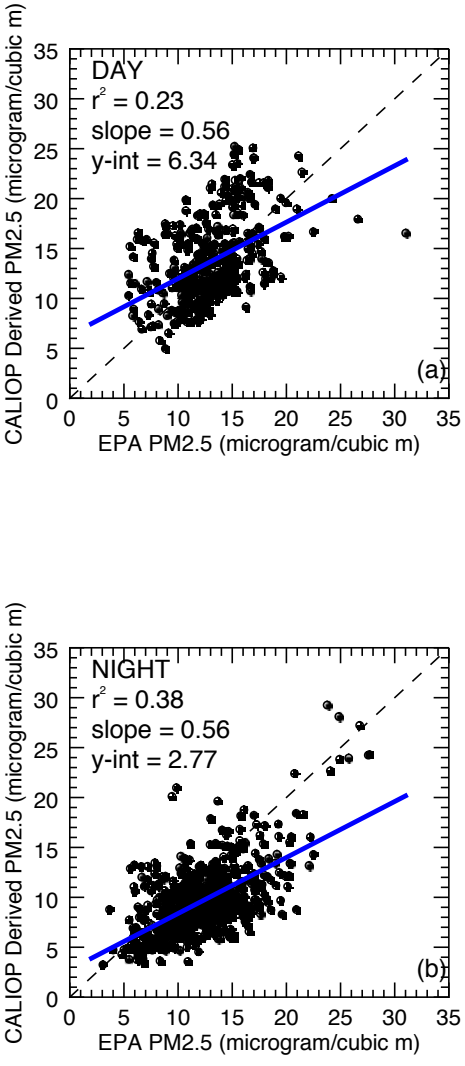

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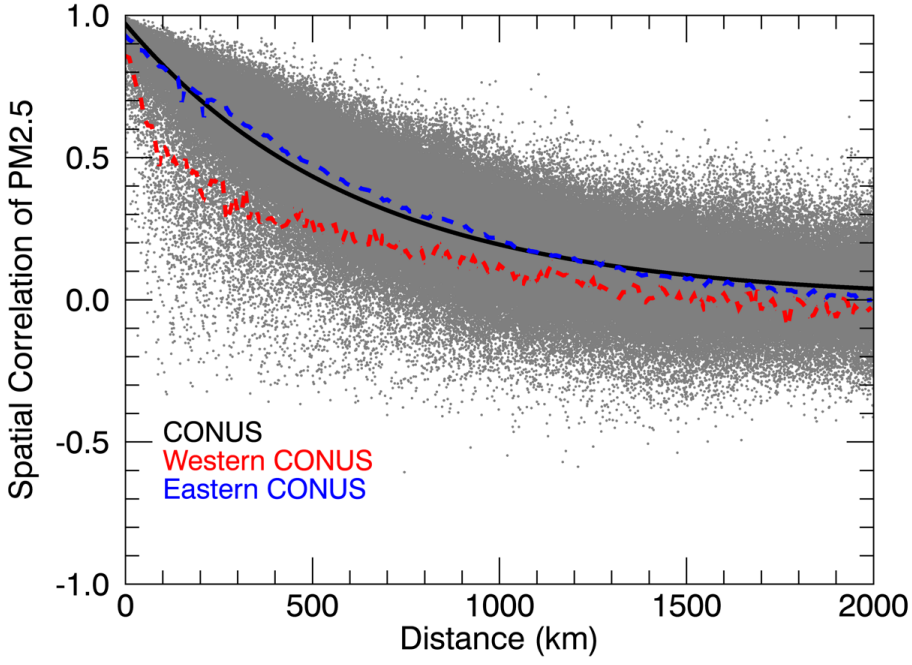

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888 **Tables**
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Parameter	Values
Integrated_Attenuated_Backscatter_532	≤ 0.01 sr^{-1}
Extinction_Coefficient_532	≥ 0 and ≤ 1.25 km^{-1}
Extinction_QC_532	= 0, 1, 2, 16, or 18
CAD_Score	≥ -100 and ≤ -20
Extinction_Coefficient_Uncertainty_532	≤ 10 km^{-1}
Atmospheric_Volume_Description (Bits 1-3)	= 3
Atmospheric_Volume_Description (Bits 10-12)	$\neq 0$

Table 1. The parameters, and corresponding values, used to quality assure the CALIOP aerosol extinction profile.

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Surface Layer (m)	Analysis (Day/Night)			
	R^2	Slope	Mean Bias (CALIOP - EPA; µg m^{-3})	Error Change (%)
0-100	0.27/0.41	0.69/0.38	-2.67/-9.06	-13.71/-61.94
0-200	0.33/0.53	0.77/0.75	-0.52/-5.68	3.79/-23.58
0-300	0.35/0.54	0.78/0.82	-0.09/-4.70	7.24/-12.15
0-400	0.38/0.57	0.80/0.85	-0.13/-4.25	6.92/-6.46
0-500	0.35/0.52	0.75/0.76	-0.21/-4.04	5.70/-4.39
0-600	0.40/0.53	0.76/0.75	-0.46/-3.91	3.72/-2.15
0-700	0.44/0.46	0.80/0.66	-0.41/-3.89	2.73/-2.88
0-800	0.35/0.50	0.62/0.66	-0.59/-3.76	-0.77/-2.04
0-900	0.17/0.49	0.43/0.63	-0.74/-3.74	-3.91/-2.25
0-1000	0.13/0.48	0.35/0.62	-1.08/-3.74	-7.48/-2.57
100-500	0.34/0.44	0.72/0.66	0.54/-3.40	14.21/-0.84
100-1000	**0.21/0.48**	**0.48/0.67**	**-0.39/-3.34**	

Table 2. Statistical summary of a sensitivity analysis varying the height of the surface layer, including R^2, slope, mean bias (CALIOP - EPA) of $PM_{2.5}$ in µg m^{-3}, and percent error change in derived $PM_{2.5}$, defined as: ((mean new $PM_{2.5}$ – mean original $PM_{2.5}$)/mean original $PM_{2.5}$)*100. The row in bold represents the results shown in the remainder of the paper.

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		Analysis (Day/Night)	
$PM_{2.5}/PM_{10}$ Ratio	**Slope**	**Mean Bias (CALIOP - EPA; µg m^{-3})**	**% Error Change**
Low ratio (-1 STDEV) = 0.24	0.19/0.27	-7.81/-8.61	-60.00%/-60.00%
High ratio (+1 STDEV) = 0.88	0.71/0.98	5.39/0.77	46.67%/46.67%
0.6	**0.48/0.67**	**-0.39/-3.34**	

Table 3. Statistical summary of a sensitivity analysis varying the $PM_{2.5}$ to PM_{10} ratio used, including slope, mean bias (CALIOP - EPA) of $PM_{2.5}$ in µg m^{-3}, and percent error change in derived $PM_{2.5}$, defined as: ((mean new $PM_{2.5}$ – mean original $PM_{2.5}$)/mean original $PM_{2.5}$)*100. The row in bold represents the results shown in the remainder of the paper.

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Analysis (Day/Night)							
Aerosol Type				**R^2**	**Slope**	**Mean Bias (CALIOP-EPA; µg m^{-3})**	**% Error Change**
	a_{scat}	a_{abs}	Γ				
Smoke	5.26	0.26	0.18	0.10/0.44	0.27/0.52	-1.81/-4.26	-11.53/-10.54
Sea salt	1.42	0.01	0.46	0.18/0.48	1.22/1.82	22.42/12.93	184.12/184.99
Dust	0.52	0.08	0	0.05/0.39	2.06/5.12	102.04/70.82	826.94/843.33
Sulfate	**3.4**	**0.37**	**0.63**	**0.21/0.48**	**0.48/0.67**	**-0.39/-3.34**	

Table 4. Statistical summary of a sensitivity analysis varying the aerosol type assumed in the derivation of $PM_{2.5}$, including R^2, slope, mean bias (CALIOP - EPA) of $PM_{2.5}$ in µg m^{-3}, and percent error change in derived $PM_{2.5}$, defined as: ((mean new $PM_{2.5}$ – mean original $PM_{2.5}$)/mean original $PM_{2.5}$)*100. The row in bold represents the results shown in the remainder of the paper.