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Atmospheric Measurement Techniques Discussions

Interactive comment on "A bulk-mass-modeling-based method for retrieving Particulate Matter Pollution using CALIOP observations" *by* Travis D. Toth et al.

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Response to Anonymous Referee #2

Comment: The scope of the submitted work is to investigate the potential exploitation of CALIOP extinction profiles in order to derive near-surface concentrations of particles with aerodynamic diameter less than 2.5 μm (PM2.5). The assessment of the applied methodology is made through the evaluation of the CALIOP derived PM concentrations against corresponding daily ground-based measurements obtained at numerous EPA stations, over the period 2008-2009, distributed across CONUS, which is the area of interest. A powerful element of using vertically resolved retrievals is that the altitude

range can be constrained (i.e., near surface where the PM concentrations are measured from the ground) in contrast to passive sensors which are representative for the whole atmospheric column. To my opinion, the issues addressed by the authors fit well to the scientific objectives of AMT and therefore I recommend the submitted manuscript to be published. Nevertheless, I believe that several points must be modified making the text acceptable for publication. My major and minor comments are listed below.

Response: Thank you for your thoughts and encouraging comments.

Comment: The authors have used only 2-year satellite data thus making the robustness of the obtained outcomes questionable taking into account CALIOP's low sampling frequency and narrow footprint. In order to overcome this drawback, you have to repeat the analysis for the full dataset.

Response: We agree that overcoming this sampling drawback can be achieved through extending the analysis for more than two years. However, this would be computationally expensive and is a non-trivial task. We envisioned this manuscript as a proof-of-concept study, the purpose of which is to provide an initial demonstration of the feasibility of our method. Adding other years to the analysis will be one of the focuses of forthcoming CALIOP/PM2.5 papers.

Comment: According to the applied methodology, all the aerosol extinctions assigned as dust in the CALIOP retrieval algorithm are masked out since focus is given on the small size particles (Lines 198-200). However, which is the treatment for the other aerosol subtypes consisting of coarse particles (i.e., marine, dusty marine)? Moreover, what is happening when the aerosol subtype is clean continental? I would suggest to repeat the aerosol type analysis (Section 3.2.8) but considering only the CALIOP aerosol subtypes which are not associated with large size particles (i.e., dust, marine, marine dust) and are relevant to pollution. Keep in mind that appropriate modifications, depending on aerosol types, may be needed in equations 1, 2 and 3 (i.e., mass scattering and absorption efficiencies, hygroscopic growth factor).

Response: All aerosol subtypes not classified as dust are considered for our method (e.g., marine, dusty marine, clean continental, etc.). We have already excluded dust, and most areas of the CONUS are not dominated by sea salt aerosols. Indeed, a statistical analysis showed that CALIOP 100-1000 m aerosol layers consisting entirely of marine (dusty marine) subtypes represent only $\sim$2% ($\sim$1%) of all subtypes. Thus, the impact of including these aerosols should be minor. We do note, however, that one of the areas of focus for future studies of CALIOP-derived estimates is a more thorough investigation into aerosol typing.

Comment: Could you please comment why the quality assurance criteria applied here are different than those suggested by Tacket et al. (2018; https://www.atmos-meas-tech.net/11/4129/2018/)?

Response: The QA criteria applied for this paper are the same as those of our previous CALIOP papers (e.g., Toth et al. 2014; 2016; 2018), and we wanted to be consistent with these studies. The QA scheme employed here was developed from Kittaka et al. (2011) and Campbell et al. (2012), both of which provide detailed justifications for the QA choices made. Toth et al. (2016) provides comparisons of aerosol extinction profiles using our QA scheme and those from the CALIPSO Level 3 aerosol profile product. While some differences were found, these were mostly attributed to differences in averaging, treatment of clouds and fill values, and our vertical regridding from 60 m to 100 m.

Comment: Page 7 – Lines 157-160: The inclusion of different PM measurements techniques (filter-based or averages from hourly samples) how can affect the intercomparison results?

Response: As suggested by Kiss et al. (2017), large uncertainties exist in hourly PM data, while less biases are expected for PM data collected over a longer period of time. Thus, there are likely differences in the two methods for collecting PM data. Still, to fully explore this issue requires a study of its own, and thus we have added the following

discussions in the text:

"Note that uncertainties have been reported for hourly PM measurements (Kiss et al., 2017). Examples of some uncertainties in these PM2.5 measurements depend upon the instrument/method used: gravimetric (e.g., transport to the lab/human error and volatization of PM during the drying process; Patashnick et al., 2001), TEOM (e.g., errors due to improper inlet tube temperature; Eatough et al., 2003), and beta attenuation monitors (e.g., changes in the sample flow rate due to variations in temperature and moisture; Spagnolo, 1989). Also, it has been found that beta attenuation monitors may be more accurate than TEOM, as TEOM can underestimate PM2.5 at low temperatures (e.g., Chung et al., 2001). Still, as suggested by Kiss et al. (2017), PM data collected over a longer period of time are much less likely to be biased. Thus, we expect lower uncertainties from data collected over 24-hours, then daily data generated by averaging hourly observations. Fully quantifying the differences from the two different PM observing methods, however, is the subject for a future study."

Comment: Page 4 – Lines 97-102: How much reliable are the scatterplot metrics when MODIS provides daylight AODs while PM concentrations are daily averages? Have you noticed any variation both in spatial and temporal terms?

Response: In this paper, we have not looked into the spatial/temporal variations of MODIS AOD versus PM2.5. This, however, was the subject of one of our past studies (Toth et al., 2014), for which MODIS AOD was compared to both daily (within 1x1 deg.) and hourly (within 40 km) PM2.5 measurements. While larger correlation coefficients were found for the hourly analysis, they still remained low. The purpose of Fig. 1 in this paper was to simply illustrate the limitation of using column-integrated AOD from passive sensors to estimate PM2.5 concentrations near the surface.

Comment: Page 9 – Line 202: A couple of citations are needed here in order to support this argument.

Response: We have added two citations (Nessler et al., 2005 and Lynch et al., 2016),

as requested.

Comment: Page 10 – Lines 236-238: It will be useful to provide a map with the number of days participating for the calculation of the average maps illustrated in Figure 3. Moreover, it is required a geographical distribution providing the average number of profiles considered for the derivation of 1° x 1° grid cells (i.e. an indicator of spatial representativeness within the 1deg grid cell).

Response: Thank you for this suggestion. We have added the requested maps as a figure in an appendix. Also, the following description was added to the text in Section 3.1: "Note that, for context, maps of the number of days and CALIOP Level 2 5 km aerosol profiles used in the creation of Fig. 3a-d are shown in Appendix Fig. 1."

Comment: Page 12 – Lines 270-279: I don't agree with the collocation criteria applied here. The horizontal distance (100 km) between CALIOP and PM station probably is too long since the analysis focuses on PM2.5 originating from pollution. Under these cases it is expected that the horizontal variability will be very strong and the concentrations will decrease rapidly for increasing distance from the source. As it concerns the temporal collocation, the optimum solution would be to use PM measurements available at the finest temporal resolution thus making feasible an appropriate matching with the CALIOP near-surface profiles. On the contrary, if the ground-based data are provided only as daily averages then you cannot consider that a satellite overpass and a daily average are temporally collocated. In the former data you have an instantaneous observation while in the latter one the diurnal variation is included. In case where the EPA data are given only on a daily basis, then it is more convenient to compare "daily" CALIOP profiles (considering dates where both the daytime and nighttime satellite retrievals are available) against the corresponding surface PM10 concentrations. For this reason, I believe that Figures 3-e and 3-f as well as the relevant parts of the text must be removed. Please consider this comment throughout your analysis.

Response: Thanks for the suggestion. We agree that "daily" averaged CALIOP profiles

may be used for comparing with daily averaged surface PM observations. However, with a narrow swath of $\sim$70 m and a repeat cycle of 16 days, very few data points would be available within 100 km of a particular EPA site for both daytime and nighttime CALIOP aerosol profiles. For the spatial collocation, the +/- 100 km collocation distance is used here, as we considered the spread of aerosols within 24 hours. For example, for a 10 km/hour wind speed, aerosol particles may travel 200 km (or +/- 100 km) within 24 hours. Also, as suggested from this paper, the averaged e-folding correlation length for PM2.5 concentrations over the CONUS is $\sim$600 km, and thus we believe 100 km is a reasonable collocation range.

Also, analysis using finer temporal resolution PM2.5 data may produce better results under some conditions, but comes with its own issues. For example, there are insufficient collocated CALIOP profiles and hourly PM2.5 data over a two-year period for the CONUS, so the temporal domain would need to be greatly expanded. Secondly, this type of study would take careful analysis of the CALIOP data, as individual CALIOP aerosol extinction profiles could be subject to higher uncertainties (e.g., rather than using a two-year mean). These research topics will be examined in detail in future studies.

Comment: Section 3.2.1: Considering my previous comment, the analysis should be presented only for the "daily" CALIOP – PM pairs and not separately for daytime and nighttime. Likewise, the CALIOP derived PM2.5 ranges (x axis in Figure 5) should be equally sampled and not grouped based on user-defined bins of PM concentrations. In addition, the authors are stating in Lines 314-316 that the computations have not been done for PM concentrations $\geq$ 25 μg m-3 due to the limited number of concurrent annual means. However, according to Figure 5, the number of samples for the lowest bin (< 5 μg m-3) during daytime is almost zero (the same is valid for the highest bins, particularly for the nighttime retrievals). Is that correct? Can we trust the calculated RMSEs resulting from a very small number of samples?

Response: As mentioned in another response, using only "daily" CALIOP-PM pairs

is not feasible here for a robust analysis, due to the repeat cycle of the CALIPSO satellite. Very few data points would be available within 100 km of a particular EPA site for both daytime and nighttime CALIOP aerosol profiles. Thus, we leave daytime and nighttime separated for this figure. As for the CALIOP derived PM2.5 ranges, we have adjusted them such that each bin is equally sampled based upon a cumulative histogram analysis. Each point from left to right in the new Fig. 5 represents the RMSE and mean PM2.5 concentration derived from CALIOP for 0-20%, 20-40%, 40-60%, 60-80%, and 80-100% cumulative frequencies. This addresses the other items in this comment, like those of few samples for the lowest and highest bins in the old Fig. 5. Because they are now equally sampled, we have removed the secondary y-axis since the number of samples do not change as a function of CALIOP-derived PM2.5 concentration.

We have also revised the corresponding text in Section 3.2.1 as follows:

"As a first step for the uncertainty analysis, we estimated the prognostic error of 2-year averaged PM2.5_CALIOP. Figure 5 shows the root-mean-square error (RMSE) of CALIOP-based PM2.5 concentrations against those from EPA stations as a function of CALIOP-based PM2.5 for the 2008-2009 period over the CONUS. RMSEs were computed for five equally sampled bins, determined from a cumulative histogram analysis. Each point in Fig. 5, from left to right, represents the RMSE and mean PM2.5 concentration derived from CALIOP for 0-20%, 20-40%, 40-60%, 60-80%, and 80-100% cumulative frequencies. A mean combined daytime and nighttime RMSE of $\sim$4 μg m-3 is found, with a mean value slightly greater for nighttime ($\sim$4.3 μg m-3) than daytime ($\sim$3.7 μg m-3). While most bins exhibit larger nighttime RMSEs, daytime RMSEs are larger for the greatest mean CALIOP-derived PM2.5 concentrations."

Comment: Section 3.2.2: To my opinion this sensitivity study should be the first step of the analysis in order to define the most "representative" altitude range. According to the summary statistics presented in Table 2, it seems that it is better to restrict the upper bound at 600 – 700m.

Response: While a surface layer up to about 600-700 m results in larger r2 values, much variability in the statistics exists between surface layer heights (as shown in Table 2). Also, differences are found between daytime and nighttime for various layers. One possible issue is a lower signal-to-noise ratio if we restricted the surface layer to lower heights. We stress that the purpose of this paper is an initial exploration of the topic, and wanted to include Table 2 as a first look at surface layer height sensitivity. Another study is necessary to better evaluate this subject, especially as surface layer height changes regionally and diurnally.

Comment: Section 3.2.4: Which is the impact on the r2 values?

Response: The r2 values are not impacted by varying the PM2.5/PM10 ratio. This is because all of the CALIOP-derived PM2.5 points for each scenario shown in Table 3 are multiplied by a common ratio (see Equation 3), but the collocated EPA concentrations remain unchanged (thus not altering the correlation).

Comment: Section 3.2.5: Instead of presenting daytime and nighttime CALIOP derived PM concentrations it is better to consider only the daily (computed from the concurrent daytime and nighttime profiles) ones (see comment 6).

Response: Thank you for the suggestion. We believe it is important to show the daytime and nighttime analyses separately, and an analysis using concurrent daytime and nighttime profiles collocated with a particular EPA site will not yield many samples due to the repeat cycle of the CALIPSO satellite. Thus we didn't make the change.

Comment: Page 19 – Lines 448-450: This means that the CALIOP derived PM concentrations are not reliable in coastal (contamination by sea-salt particles) or dust affected regions?

Response: The large uncertainties are because mass extinction efficiencies are drastically different for coarse and fine mode aerosols. Here we applied mass extinction efficiencies from fine mode aerosols to coarse mode aerosols, and not surprisingly,

see large uncertainties. Lower uncertainties can be expected if we apply coarse mode mass extinction efficiencies to coarse mode aerosols. However, this puts the pressure on accurate estimations of aerosol types from CALIOP or other lidar observations, which we believe is a study of its own, and will be investigated in future studies.

Comment: Section 3.2.9: In this section it would be also useful to provide a map with the distances where the 1/e value is found at each station.

Response: Thank you for this suggestion. However, for each pair of PM observing locations, one correlation value is computed for a given distance between the two locations. Thus, the analysis is discrete, not continuous. The 1/e values are estimated from Fig. 10, which is composed of individual points representing both a distance and spatial PM2.5 correlation between pairs of EPA sites over the CONUS. If we apply the same analysis to a given PM observing location, it is likely to have data gaps due to the discrete nature of the dataset. Thus, we leave Fig. 10 untouched.

Comment: Page 3 – Lines 81-84: Could you please explain better this sentence?

Response: We have rewritten the sentence to:

"Indeed, Kaku et al. (2018) recently showed that surface PM2.5 had longer spatial correlation lengths than AOD, even in the "well behaved" southeastern United States where previous studies showed good correlation between PM2.5 and AOD (e.g., Wang and Christopher, 2003)."

Comment: Page 4 – Lines 91-94: It is not clear what the authors want to say here.

Response: We have rewritten the sentence to:

"It is arguable that from a climatological/long-term average perspective, the use of AOD as a proxy for PM2.5 concentrations nevertheless has certain qualitative skill (e.g., Toth et al., 2014; Reid et al., 2017) due to the averaging process that suppresses sporadic aerosol events with highly variable vertical distributions."

Comment: Page 10 – Line 244: What do you mean exactly here? ("..., as surface layer heights may change seasonally and diurnally.")

Response: We have removed "as surface layer heights may change seasonally and diurnally" to avoid confusion.

Comment: Page 19 – Line 431: Sulfate & organic or just sulfate?

Response: To avoid confusion, we removed "& organic". But primary and secondary biogenic aerosols are mostly fine mode as well.

Comment: Page 20 – Lines 456-458: Please rephrase this sentence.

Response: This sentence was broken into two sentences, as follows: "To accomplish this, all EPA stations over the CONUS with at least 50 days of daily data available for the 2008-2009 period were first determined. Next, the distances between each pair of these EPA stations, and their corresponding correlation of daily PM2.5 concentrations, were computed."

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