Reply to Reviewer 1

This study evaluates the uncertainties associated with geophysical approaches to derive surface $PM_{2.5}$, based on satellite AOD and modeled $PM_{2.5}/AOD$. The authors go through a very detailed evaluation of all the potential factors, using ground-based observations of $PM_{2.5}$, AOD, aircraft observations of aerosol extinctions/composition, and atmospheric soundings of RH over the Northeast United States. The analysis is very comprehensive, the paper is well written and I commend the authors for presenting the results in a succinct way on the figures.

Reply: We would like to thank the reviewers for their time and effort to review our manuscript. We have revised the manuscript following the reviewers' suggestions.

One suggestion that I have for the authors is to present a figure with time series of the daily variations in $PM_{2.5}$, AOD, and $PM_{2.5}$ /AOD. The manuscript only contains barplots of the biases and Pearson correlation coefficients, and there would be value for the reader to see the actual timeseries. I found Figure 1 very interesting in terms of displaying the contributions of different factors to spatial variability in satellite-derived $PM_{2.5}$. Something similar to illustrate the controlling factors for the daily variability would be valueble.

Reply:

That is a great point. We added a figure showing the temporal variability of regional average AOD_{MAIAC} , $PM_{2.5_CMAQ}/AOD_{CMAQ}$, and $PM_{2.5_MAIAC}$ (Figure 2). Similar to Figure 1, we show that the temporal variability of $PM_{2.5_MAIAC}$ is mainly driven by the variability in $PM_{2.5_CMAQ}/AOD_{CMAQ}$.

We have added the following discussions in the revised manuscript:

The temporal variability in $PM_{2.5_MAIAC}$ is also mainly driven by variability in $PM_{2.5_CMAQ}/AOD_{CMAQ}$ (R = 0.61), with little temporal correlation between regional average AOD_{MAIAC} and $PM_{2.5_MAIAC}$ (R = 0.05, Fig. 2). At short time scales, the daily variability in regional average $PM_{2.5_MAIAC}$ shows stronger correlation with $PM_{2.5_CMAQ}/AOD_{CMAQ}$ in all seasons except for JJA, when $PM_{2.5_MAIAC}$ are driven by variability in both AOD_{MAIAC} (R = 0.5) and $PM_{2.5_CMAQ}/AOD_{CMAQ}$ (R = 0.4, Fig. 2). Summertime AOD_{MAIAC} is higher than wintertime AOD by 50%, while summertime $PM_{2.5_MAIAC}$ is lower than in winter by 46%. Previous studies also found inconsistent seasonal cycles in AOD and $PM_{2.5}$ (Ford et al., 2013; Kim et al., 2015). We attribute the opposite seasonal cycle in $PM_{2.5_MAIAC}$ and AOD_{MAIAC} to three factors: 1) weak boundary layer ventilation in winter that leads to sharp vertical gradients of aerosol distribution (Kim et al., 2015); 2) higher RH in summer that leads to larger hygroscopic growth; 3) model

overestimates of $PM_{2.5}$ (especially OC) in wintertime and underestimates of $PM_{2.5}$ in summertime, leading to an overestimate of the winter-to-summer decrease in $PM_{2.5 CMAQ}/AOD_{CMAQ}$ (see section 3.3).



Figure 2 Regional 10-day running average of (a) MAIAC AOD (AOD_{MAIAC}, blue); (b) CMAQ modeled $PM_{2.5}/AOD$ relationship ($PM_{2.5_CMAQ}/AOD_{CMAQ}$, red); and (c) satellite derived $PM_{2.5}$ ($PM_{2.5_MAIAC}$, green). The numbers on the upper left corner show the Pearson correlation coefficients (R) of $PM_{2.5_MAIAC}$ with $PM_{2.5_CMAQ}/AOD_{CMAQ}$ (red) and AOD_{MAIAC} (blue).

References:

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