Review of "10 yr spatial and temporal trends of PM2.5 concentrations in the southeastern US estimated using high-resolution satellite data" by X. Hu et al.

This paper shows the PM2.5 concentrations from 2001 to 2010 over an area in the SE U.S. and Atlanta metropolitan area using the MODIS MAIAC 1-km AOD data and a two-stage model that derives the surface PM2.5 concentrations from the AOD with a series of fitting parameters accounting for the meteorological fields, surface categories, and point emissions. The objectives are (1) to estimate PM2.5 concentrations in the study domain with MAIAC AOD as the primary predictor and other variables as the secondary predictors, (2) to generate maps of annual mean PM2.5 concentrations from 2001 to 2010, (3) to examine the 10-year temporal trends of PM2.5 in the study domain and Atlanta metro area, and (4) to investigate the potential impact of fires on PM2.5 levels.

While it is valuable to use the high-resolution satellite AOD data for PM2.5 prediction and trend analysis, this paper has not shown the unique value of using such data and does not provide quantitative assessment on the connection between emission and PM2.5 level. My major comments are listed below. Major revision and a great deal of clarification are necessary before the paper can be considered for publication on ACP.

1. The use of 1-km resolution AOD data: I found that the paper has not demonstrated the merit of using high-resolution satellite data. Although it is stated in the "Introduction" that the standard MODIS and MISR products at 10-km and 17.6-km, respectively, are not sufficient for epidemiological studies and omit details of PM2.5 spatial variability, the results and analysis presented in this paper do not show any advantage of using the 1-km AOD data other than the visual structure in the maps. Considering that most analysis presented in the paper was done based on large area averages (either the study domain or Atlanta metro), what can the 1-km AOD data offer but the 10-km data cannot for the purpose of the present study? What is the spatial scale of AOD or PM2.5 variability that makes 10-km data insufficient?
2. The two-stage model: This model depends on a large number of fitting parameters. It is not clear, however, from equations (1) and (2), how these parameters are obtained or derived. Did you use the observed PM2.5 and AOD plus other data to construct all the bi? What are the random and fixed intercept and slopes, by definition? Do different meteorological fields (winds, PBL, RH, etc.) associated with different b2s? How do road length associate with the site point location of individual site? Are all these coefficients "day-specific"? Among the secondary predictors, which ones matter the most? How are these secondary predictors chosen? The model and the methods are not clearly presented in the paper and clarification is necessary, especially there is no previous publication or documentation that might serve as a reference for the method.
3. Model fitting: It is said that "the model was fitted for each year individually" such that the predictors may vary for different years. I wonder why it was not fitted for each season, instead of for each year, since the seasonal variations of aerosol and meteorological variables are much stronger than the interannual variations, so doing seasonal fitting makes more sense.
4. Error and uncertainty: There is no estimate of the range of error or uncertainty in this method, especially so many empirical fitting parameters have been used. It seems that

aerosols above the PBL is not considered at all. Even though most time aerosols may be indeed concentrated in the PBL in the study region, such omission should be discussed.

5. PM2.5 trend and the cause of the decreasing trend: It is obvious from Fig. 3, 4, 5, and 7 that the PM2.5 started to drop in 2008. Before that there was just small fluctuations. There is no "generally decreasing trend" during the 10-year period; rather, it looks like a step function with a significant change occurring in 2008. What causes such change, however, is not adequately analyzed. It is mentioned a few times in the paper that the reduction of PM2.5 "might be due to recently enacted emission reduction program", "is probably due to dramatically reduced number of emission sources", etc., the quantitative relationship between emission and PM2.5 is not presented at all. I wonder why more quantitative analysis was not done, as the point emission is actually one of the variables used in the two-stage model on at least yearly basis, so the authors must have access to the emission data for all these years to see the year to year emission changes and link them to the PM2.5 changes.
6. Impact of fire emission on PM2.5 level: This part of the study is particularly weak – basically there is no quantitative analysis of the fire impact. The only display that may suggest some fire impact is the difference of fire occurrence and PM2.5 levels between the two rural sites showing some coincidental peaks and valleys. Why is it necessary to show the difference between the two sites, instead of showing the variation of PM2.5 level and fire occurrence at the sites affected by the fire? Even if you choose to use the difference between the two sites, can you be more quantitative, e.g., make a scatter plot of the delta_PM2.5 vs. delta_fire? Is your study consistent or different from Zhang et al. 2010 that shows 13% PM2.5 in the SE U.S. is from fire?

Other comments:

P 25618, line 8-9: "inherent disadvantage…". But you really have not demonstrated such a disadvantage for your study. Also, what AOD products are considered as "current"? MODIS currently has 3-km product. MAIAC is also a current product.

P 25618, line 11-13, MAIAC: MAIAC is one of the MODIS products, which retrieves AOD from MODIS measurements using the MAIAC algorithm. This should be clarified to not mislead readers as MAIAC is an independent AOD product from a different sensor.

P 25620, line 12-13, and line 25: Again, this is about the "coarse" resolution product: Can you elaborate why 10- or 17.6-km product cannot serve your purpose? What is the aerosol spatial variability that determines the adequacy of product resolution?

P 25621, line 12: It sounds like you have more than one objective. The paragraph should be re-phrased.

P 25623, line 12-13: What differences it may introduce from using just Terra, just Aqua, or both Terra and Aqua?

P 25623, line 14-15: A combined use of AOD at 10:30 am and 1:30 pm can only produce the estimated PM2.5 averaged at these two particular time, not "between 10 am to 2 pm".

P. 25625, equation (1): As I mentioned earlier, this equation needs to be better explained.

P. 25625, line 20: What are the definitions of “fixed and random intercept and slopes” and how are they obtained?

P. 25626, line 1-13: “may include” – what are actually included? Do different met fields associated with different b0 and b2 values? It is hard to understand from eqn. (1). Maybe a detailed description (can be in a form of Appendix or Supplemental Material) is necessary if this method has not published in the literature. Are other land cover types considered other than forest cover?

P. 25627, line 3-4: the sentence “That is,…” is confusing.

P. 25627, line 15-16: As mentioned earlier, I don’t understand why the fittings are done for each year individually, not for each season (or month).

P. 25628, line 3-4: meteorological fields should have much stronger day-to-day, month-to-month, or season-to-season variations than year-to-year variations.

P. 25628, line 26: “…occur in the south of the study domain”: From Figure 3 the high PM2.5 is the SE triangle area in the study domain, not “south”.

P. 25869, line 1: What is the magnitude of the agriculture emission? Does it comparable with the urban industrial emission? Is the ag emission part of your predictors in equation (1)?

P. 25869, line 5-6: Biomass burning emission is very seasonal. You should look the seasonal maps.

P. 25869, line 7: “corresponds well” – what is the criteria of “well”? Should have a quantitative measure instead of a subjective phrase.

P. 25869, line 10, Fig. 5: What is the last panel in Fig. 5 that is never discussed?

P. 25629, line 16, “percent changes” - How do you obtain the % change? By linear fit of the time series, or by the difference between 2010 and 2001? It is said the change is “from 2001 to 2010” but on the next page it is said “between 2001 and 2010”. Please clarify how the changes are calculated.

P. 25630, line 1-15: The relationship between emission and PM2.5 should be better analyzed. As I mentioned at the beginning, if the two-stage model considers the emission as a predictor of PM2.5 concentration, why can’t you pull out the emissions of each year to see if the increase or decrease is indeed due to the emission changes, and if they are of similar magnitudes?

P. 25630-25631, section 3.4: Analysis in this section is too descriptive. More quantitative assessment is necessary. (a) Also there is no general declining trends – PM2.5 is significantly lower in the last three years, but there is no steady decline from 2001 to 2010. (b) For comparisons with the observation at the monitoring sites, you should compare your results with the obs at the same sites. Although you want to look at the trend at larger area, you could have shown the site comparisons on the same figure, maybe with a dotted line. (c)

Again, do you have the emission to support your claim that the increase of sulfate due to the higher emissions from electric utilities and industrial boilers in 2005? (d) When was the emission reduction programs enacted? Which year is "recently"?

P. 25631, section 3.5: This section does not tell us anything. Everyone expected that fire will have impact on PM2.5, so seeing some peaks and valleys of PM2.5 change with fire activity really is not any news. It would be more useful, given you have 10-year data, to estimate the quantitative magnitude of fire contribution to PM2.5 in fire-effected sub-domain from year to year, or even using only one-year data to show some quantified analysis.

P. 25631 and Fig. 8: The fire activity certainly does not correspond to the PM2.5 changes anywhere within the study domain.

P. 25631, line 27: The estimated 13% contribution is not from the present work but from Zhang et al., 2010. It should be clarified. This study did not show any quantitative number.

P. 25632, line 8-9, the estimate at coarser scales "inevitably omit local spatial details" – but you did not use any local and spatial details in this study, so why the resolution matters?

P. 25632, last paragraph: Such statement can only be examined by comparing the PM2.5 changes over the entire domain as well as over the EPA monitoring sites.