Authors’ response to the review comments #2

Title: Evaluation of modeled surface ozone biases as function of cloud cover fraction (gmd-2015-42)

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Anonymous Referee #2

The authors describe a new method for evaluating air quality models. They present an observational constraint on the surface ozone/cloud relationship for the continental USA, using observations from the AirNow air quality network and cloud data derived from the satellite-mounted MODIS instrument. New ways to evaluate models are always welcome, and this is an interesting addition to our evaluation arsenal. Ultimately, I think that the study could be a good addition to the literature, but I feel that the authors claim too much for the method, and their conclusions should be more circumspect. It would also benefit from further statistical analysis. I have comments related to this below.

General response: The authors express their appreciation to the two reviewers and the editor. We believe that their comments are very productive and substantially contributed to improve the manuscript. We offer point-by-point responses to the issues and comments addressed by reviewers. Reviews’ comments are shown in italics. Figures 1-4 indicate figures in the new manuscript, and Figures R1-R5 indicates figures in this reply.

We thank to both reviewers for mentioning the statistical significance of current analysis. We agree that current analysis with all site data can be limited due to the high uncertainty from local characteristics of individual sites. In order to supplement current analysis, we include an additional analysis (Figure 4) for the cloud fraction (CF)-O3 correlation for each AQS monitoring sites to minimize the individual local characteristics, showing geographical distributions of CF-O3 correlation. Hopefully, this analysis can provide additional information for the issues that reviewers have commented.

As the reviewer #1 mentioned, this manuscript tries to raise a question on the CF handling which hasn’t been addressed much after the early stage of air quality model development. The estimation of CF impact to ozone bias from this manuscript is intended to provide a theoretical range of impact. We do not claim this guess is a finalized quantitative interpretation. We do understand more accurate quantity can be reached by further investigating individual site’s behavior after removing other local uncertainties (e.g. emission variation). Hopefully, we can pursue it in the following studies.

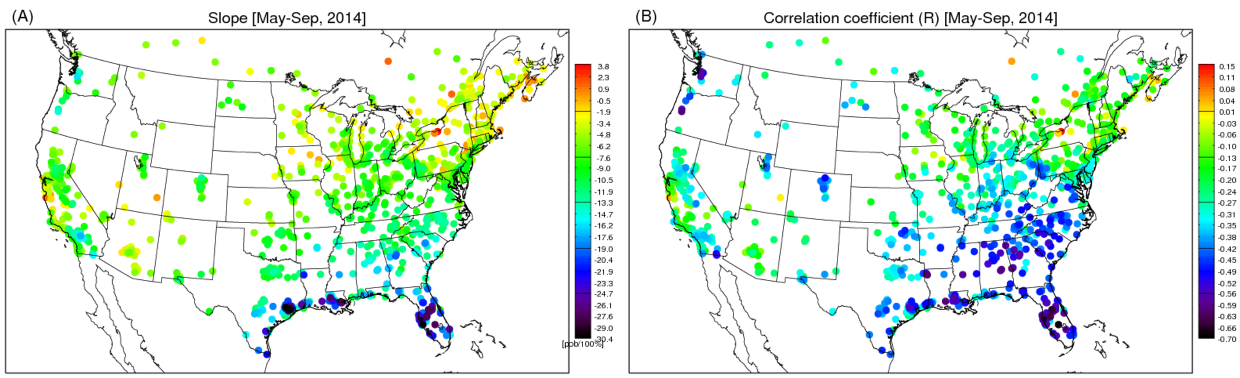

Figure R1. Spatial distributions of (a) slope and (b) correlation coefficient of linear regress between MODIS CF and MDA8 ozone.

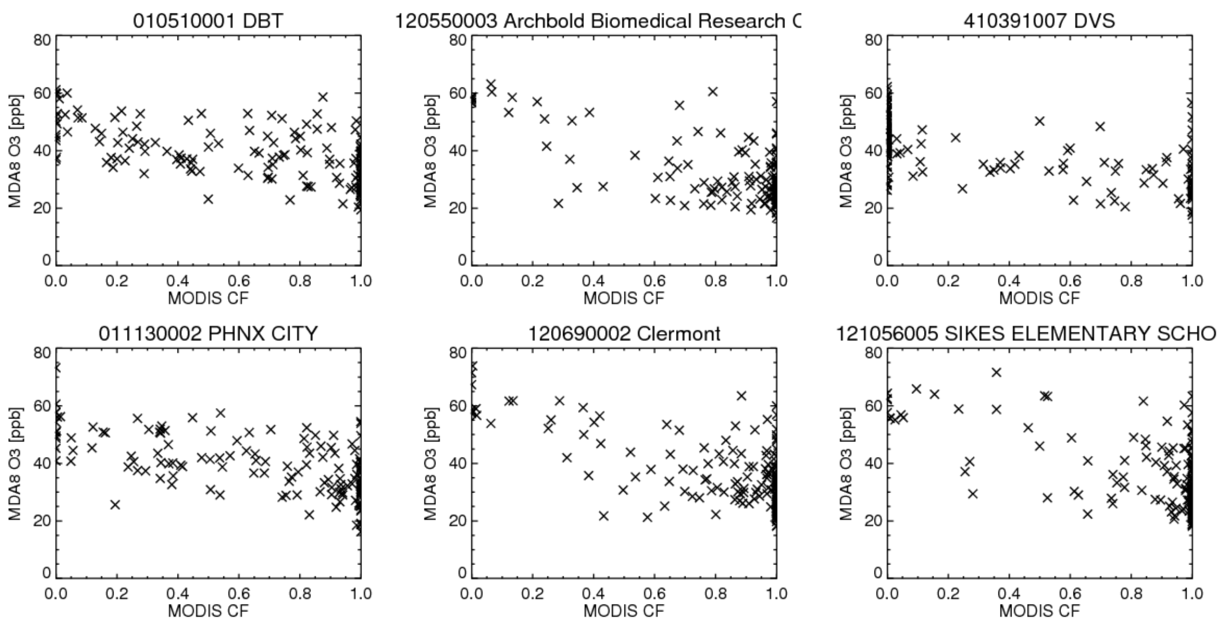

Figure R2. Scatter plots of MODIS CF & MDA8 ozone for 6 selected AQS sites.

General comments

1. Interpretation. Ozone chemistry is very complicated and depends on many things, which is something the authors themselves note in L164. As such, I don't think that attributing x% of the model bias to cloud fields can be done (L182). How can one disentangle this bias from (say) a bias in the emissions? If the emissions biased things one way, the cloud bias might correct it or intensify it. Instead I think that this technique potentially adds another useful constraint on model performance, but one that should be used in conjunction with other evaluation methods (MDA8, pdfs of monthly stats, long term climate relation-ships etc).

Reply: Thanks for the comment. We agree that biases from emissions have strong impact on the bias of surface ozone. In this manuscript, we, therefore, focused on the relative changes of ozone bias according to relative changes of CF difference, instead of absolute ozone bias values (e.g. regression slope). We agree the estimation of CF impact may be revealed more clearly if the impact from emissions is removed. We intend to further pursue it by removing emission pattern (e.g. weekly pattern) in the future study for individual monitoring sites.

In addition to these comments, there is a distinct lack of statistical rigor in the interpretation of the relationships. The authors should at least quote uncertainties on the regression coefficients for (e.g.) Figure 3 – are they in fact statistically different from zero?

Reply: We agree that Figure 3 is too noisy since it includes data from all sites. For better clarification we included individual site's analysis in Figure 4, showing its geographical distribution with better correlation, and they are showing higher correlation especially in the southern states.

Manuscript change: Additional analysis and descriptions are included; Figure 4 and line 175-186

Also, what is meant by "correlation slope"? Slope from the linear regression perhaps?

Response: Corrected.

Regarding correlations, the authors might like to see if there is a significant correlation between CF and MDA8, for both the "standard" (Pearson) correlation and a rank correlation. They will likely need to be careful in their interpretation of the significance here since, depending on spatial autocorrelation, each site will likely not represent an independent sample.

Reply: We included spatial distribution of Pearson correlation coefficients for individual sites in Figure 4. We understand each site does not totally stand along since ozone is secondary pollutant affected by local transport of emissions and precursors.

Finally, do the authors think that these relationships would be broadly applicable to other regions, or even for global models?

Reply: Thanks for the comment. This is one of our future study plans. We are trying to apply the same methodology to East Asia. Figure R5 shows preliminary results: MODIS CF & MDA8 ozone (around 300 surface monitoring sites from National Institute of Environmental Research (NIER), Korea) during May 2014 over S. Korea (left) and long-term seasonal variation of CF-Ozone sensitivity for 12 year (right). We could find very similar correlation from Korean data.

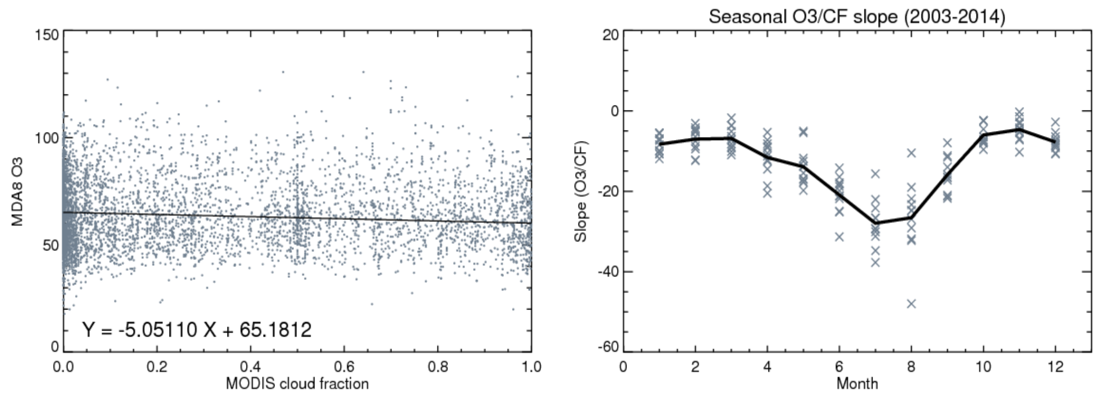

Figure R5. Scatter plot between MODIS CF and MDA8 ozone over Korea during May 20014 (left), and seasonal variations of CF-O3 sensitivity during 2003-2014 (right)

2. Introduction. I'm not sure that the introduction sets up the paper all that well: - It would be useful to mention the other techniques that are used to evaluate AQ models to give some context for this work (and something to refer to in the conclusions) – The first paragraph of the introduction talks about the importance of aerosols for photolysis rates, but my understanding is that CMAQ (in common with most other models) does not consider aerosol scattering when it is adjusting the photolysis rates. It would be a good idea to mention this I think. - The authors also might like to think about what photolytic processes are most important here: jNO2, jO3P and jO1D, or others?

Reply: Thanks for the comment. As j(NO2) (<420nm) leads to the ozone production and j(O1D) (<340nm) results in the ozone loss, the type of UV radiation reaching to the surface is important for surface ozone concentration. In general, UVA (315-399 nm) mostly reaches to the surface without absorption to the ozone layer, it has higher chance of ozone production by improving j(NO2) at the surface level, compared to the ozone loss by j(O1D). Detailed analysis, however, on the quantitative interpretation of each photochemical processes are beyond the scope of current study. We like to pursue more detailed analysis in the future. Introduction is also rewritten to address the importance of adjusted UV radiation (by cloud and aerosol) to ground level ozone. We agree that CMAQ's photolysis adjustment by aerosol is an important issue, but it is beyond this study's scope since it should be handled in the frame of in-line feedback modeling system. We believe EPA is working on this issue.

- Finally, the introduction could also mention some of the work that has looked at the potential role of clouds (through photolysis) in interannual variability of tropospheric composition (e.g. Voulgarakis et al. (2009), ACP, doi: 10.5194/acp-9-8235-2009).

Reply: Thanks for the recommendation. We cited Voulgarakie et al. (2009) in the manuscript.

L14. Is this "clear" correlation significant?

Reply: We removed "clear" from the abstract. From the additional analysis on the individual site, this negative correlation seems to be significant in southern states.

L31. "For instance. . ." before "Studies. . ."

Reply: Included

L81. Define CONUS

Reply: Replaced to Contiguous United States (CONUS)

L124. "serious" is rather vague

Reply: We included monthly mean values. They have 17% difference.

L144. "August 2014"

Reply: Corrected

L147. I'm not sure that I "readily expect" anything from the basics of ozone photochemistry. Would be good to have a citation here.

Reply: We replaced the sentence with detailed descriptions and included references. (Line 153-157)

L195. See my general comments. I'm afraid I don't think the study demonstrates how "crucial" it is

Reply: We understand review's concern, but the investigation of model's hidden bias is very important for regional air quality, especially on the State Implementation Plan (SIP) modeling and eventual emission control policy-making. We agree quantitative interpretation of this importance was not clear in the previous manuscript. In the additional analysis (Figure 4), coastal regions near the Gulf of Mexico show strong CF-O3 correlation up to -30 ppb/CF. In those regions, we usually experience quick evolution of local convective storms, which mean that prediction error for CF can be easily 100%. We believe that accurate modeling of those convective clouds is truly *crucial* in regional ozone simulation.

Thanks again for very productive comments.