Authors' response to the review comments #1

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

Authors: Hyun Cheol Kim, Pius Lee, Fong Ngan, Youhua Tang, Hye Lim You, and Li Pan

Anonymous Referee #1

General response: First of all, 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 including all site data can be limited due to geographical and local characteristics of individual sites. In order to supplement current analysis, we present the geographical distribution of cloud-fraction (CF)-O3 correlation (Figure R1, Figure 4 in the revised manuscript) for each AQS monitoring site. Examples from several selected monitoring sites are shown in Figure R2. 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 an estimated 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.

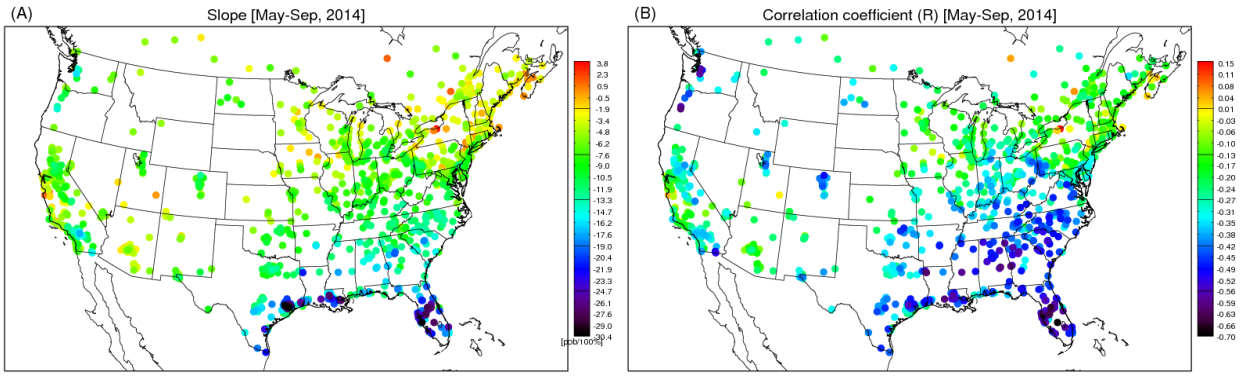

Figure R1. Spatial distributions of (a) slope and (b) correlation coefficient of linear regress between MODIS CF and MDA8 ozone.

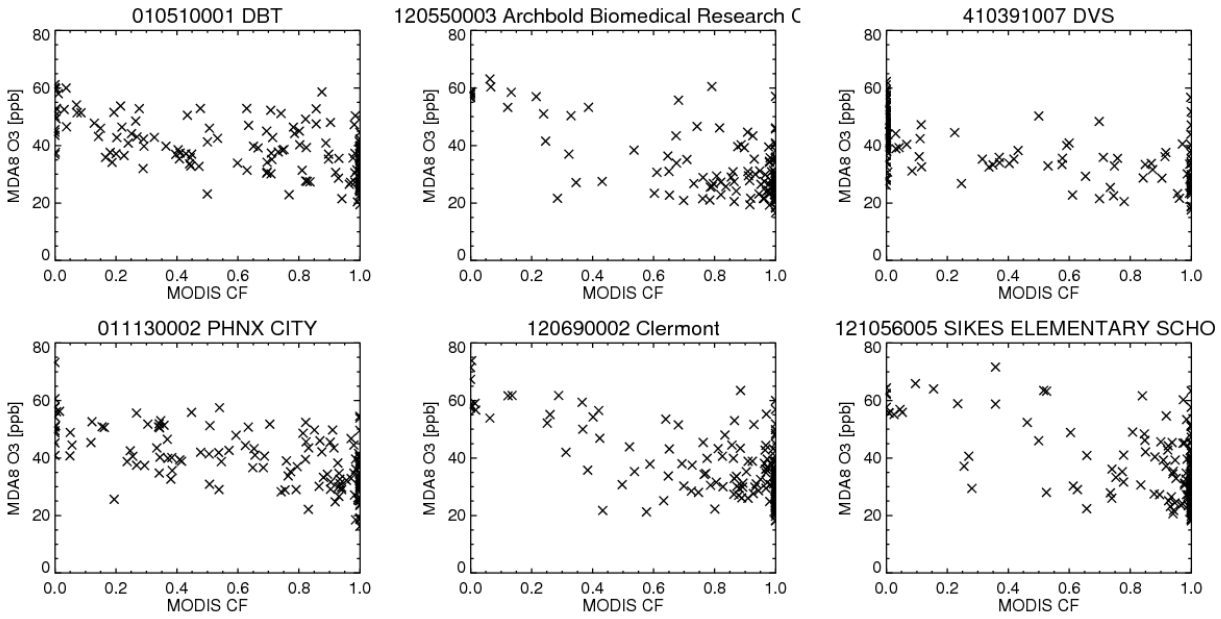

Figure R2. Scatter plots of MODIS CF & MDA8 ozone for 6 selected AQS sites.

General Comments: This paper addresses a very important issue: the source of ozone forecast bias in the NAQFC model. As this model is used extensively for operational air quality forecasting in the US, information of this type is certainly timely. The paper provides what is essentially a "back of the envelope" calculation of the impact of cloud cover on ozone forecasts. Since I have seen no prior work that addresses this issue systematically, it is a welcome addition. My main criticism is that the authors are claiming too much for their study. It can stand alone as a first rough guess calculation but probably isn't strong enough to support some of the conclusions in the paper. I think the paper is certainly worth publishing if the authors add some clarification concerning the limits of their conclusions.

Reply: Thanks for very productive comments. As mentioned in the general response, we have conducted additional analysis for each monitoring site's behavior. Also, we provide an estimation of maximum uncertainty because we intend to address that the impact of model cloudiness on O3 bias is worth further investigations.

We believe the mean of all data for each cloud fraction (CF) bin still has a meaning since local uncertainties (e.g. emission uncertainty) are independent of CF, so individual uncertainties are randomly distributed and can be averaged out. However, we strongly agree that current analysis with all site data can be limited due to the high uncertainty from local characteristics of individual sites. Hopefully, individual site analysis with better correlation will provide additional information.

Manuscript change: Additional analysis and descriptions are included – Figure 4 and line 173-184

Specific Comments:

p. 3221, line 6: re, trends in "frequency of photolysis", do you mean "rates" of photolysis?

Reply: We apologize for the confusion. It originally intended "amount of photochemical reactions". We removed this for better clarification.

p. 3222, line 15: Cloud fraction (fc) as diagnosed in the model is a function of RH, but it would be useful to know exactly what that function is as used in the experimental NAQFC.

Reply: In MCIP v3.6, cloud coverage is calculated in line 131-177 of bcldprc_ak.f90. We included it in the text.

The fractional cloud coverage used in the NAQFC is computed as following (Byun and Ching, 1999).

Cloud fraction (f_c^k) above the boundary layer:

$$f_c^k = \left[\frac{RH^k - RH_c}{1 - RH_c}\right]^2$$

Where RH^k is the relative humidity at vertical model layer k and RH_c is the critical relative humidity defined as $RH_c = 1 - 2\sigma_c(1-\sigma_c)[1 + 1.732(\sigma_c - 0.5)]$ and $\sigma_c = p^k/p^{k_{PBL}}$

Cloud fraction with the convective boundary layer when $RH > RH_c$:

$$f_c^k = 0.34\frac{RH^k - RH_c}{1 - RH_c}$$

Byun, D.W. and J.K.S. Ching, 1999: Science Algorithms of the EPA Models-3 Community Multiscale Air Qualty (CMAQ) Modeling System., EPA/600/R-99/030, U.S. EPA. Page 12-49 – 51.

Manuscript change: Added descriptions and equations (line 71-76)

p. 3224, line 2: It would be interesting to know if the changes to the experimental NAQFC noted at this line reduced the bias of the model by a magnitude that is more or less than that by cloud fractions as estimated in this paper.

Reply: We do not have sensitivity test results for August 2014. For July 2011 case, ozone overestimation is reduced from 11.44 ppb to 7.63ppb by improved model processes and emission update (33% improvement). On the other hand, current manuscript estimates 35% adjustment from 5ppb ozone overestimation by CF difference in August 2014.

Kim, H., P. Lee, L. Pan, L. Judd, D. Tong, Y. Tang, T. Chai, B. Lefer, and I. Stajner, 2014: Comprehensive comparisons of NAQFC surface and column NO2 with satellites, surface, and field campaign measurements during 2009-2014, 2014 CMAS conference, Chapel Hill, NC

p. 3224, line 6: The cloud fraction difference is estimated at 1:30 LT but the metric of interest for ozone is the 8-hour running average. This raises a few questions that probably should be addressed in the text. For example, is an instantaneous measure of cloud fraction an accurate metric with which to compare the cumulative effects of clouds and sun over an 8-hour averaging period?

Reply: As described in the manuscript, afternoon time is chosen because it is when ground-level ozone shows its highest production efficiency, which builds up its daily maximum ozone level (e.g. MDA8 ozone). We agree more CF information will help a better comparison, but it is limited since the MODIS is polar orbital. It can be further investigated with geostationary CF information (e.g. GOES data)

Is it a good measure for high, stratiform clouds more than for low level buoyancy driven clouds?

Reply: We notice cloud type (high- & low-) can be important due to the wave-length dependency of photolytic reactions. Unfortunately, current MODIS standard product does not provide separate information on the level of cloud. Our future plan for this issue is to apply an advanced MODIS cloud algorithm to investigate impacts from high- and low- clouds on the ground level chemistry. We are currently utilizing Chang and Li (2005)'s cloud retrieval algorithm to separate standard MODIS total cloud information to low-, middle- and high- clouds, but do not think results are available for current manuscript.

Fu-Lung Chang and Zhanqing Li, 2005: A Near-Global Climatology of Single-Layer and Overlapped Clouds and Their Optical Properties Retrieved from Terra/MODIS Data Using a New Algorithm. *J. Climate*, **18**, 4752–4771. doi: http://dx.doi.org/10.1175/JCLI3553.1

Is it possible that the NAQFC model "catches up" with cloud fraction as the day increases. The NAQFC, based on the NAM, uses a boundary layer parameterization scheme that may, or may not, produce low level cloudiness at the proper time in the diurnal cycle.

Reply: Since MODIS is polar orbital instrument, we are not able to fully evaluate the diurnal evolution of current model's cloud field. However, there is no evidence that current NAQFC model generates more cloud in other hour. Figure R3 shows diurnal variation of NAQFC CF and MODIS CF from current CONUS domain (land only, August 2014), showing consistent underestimation of CF from NAQFC.

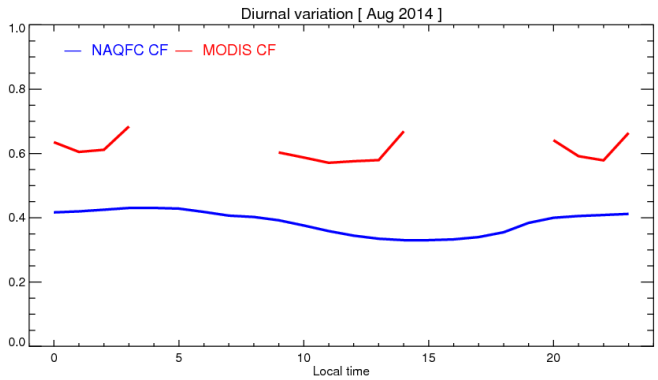

Figure R3. Diurnal variation of NAQFC CF and MODIS CF.

p. 3225, lines 20-23 and Figure 3: I didn't find this figure to be very enlightening. The text notes that Figure 3a shows "a clear separation of ground level ozone for each cloud fraction". _fig. 1 below. To be honest, I don't see much clarity in the scatterplot. Perhaps if a linear best fit line was superimposed? I find it useful for all "busy" scatterplots to include some fittted line in the figure along with the best fit equation and r and r2 values in a legend. That makes it more convenient for the reader who otherwise has to jump back a forth in the text – a table would be second best.

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. We corrected "clear separation" to moderate term. We also regenerated Figure 3 with fit line and equation.

Manuscript change: Figure 3 is replaced. Figure 4 and descriptions are included.

In any event, while the NAQFC shows less cloud cover, particularly in the near-overcast range, it is worth the effort to see whether there is a statistically significant difference between the two samples. Because we are looking at one month of data, a bit more statistical rigor would be very helpful. It's clear that cloud fraction effects are important but a little more information on the uncertainty of the estimates (that used mean values) would be very useful.

It would also be good to mention if there was anything unusual with respect to the climatology of the CONUS during August of 2014. I'm not certain that it affects the results shown here, but for the bulk of the CONUS, the summer of 2014 was a historically low ozone year – similar to 2013. As a result, the critical cases for air quality forecasts – those in the high end of the distribution (e.g., Code Orange), were scarce in 2014. _figure 2 below

Reply: Interannual variations of climatology indeed affect the base ozone level, but its sensitivity to CF in daily-timescale does not change significantly. As shown in Figure 4, the daily ozone variation due to CF change is much bigger than interannual variation, especially in southern states. To clarify this issue, we have extended analysis to 10 years (2005-2014). Seasonal variations of Ozone-to-CF sensitivity during 2005-2014 are shown in Figure R4, and they show clear seasonal variation without regard to interannual variability.

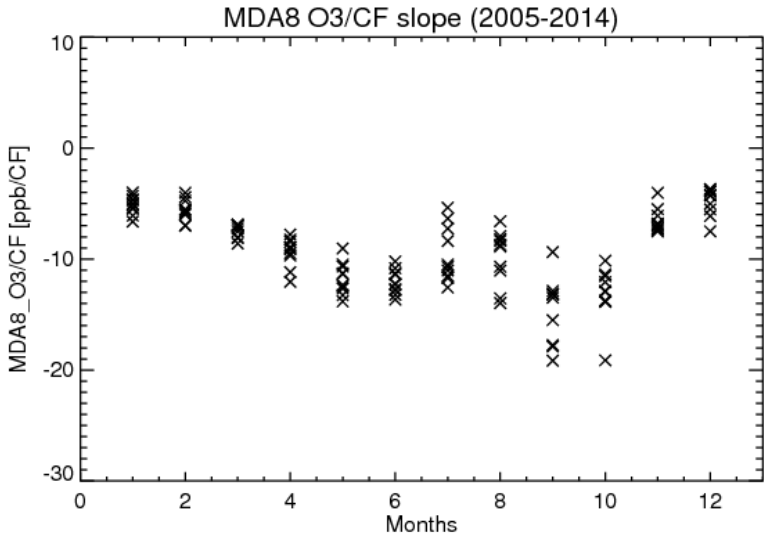

Figure R4. Seasonal variation of Ozone-to-CF sensitivity during 2005-2014.

The very clean conditions in 2014 may be a function of changing emissions but may also be associated with large scale weather patterns. See images below suggesting a cooler than normal August with large OLR anomalies (Figures via, http://www.esrl.noaa.gov/psd/data/composites/day/). _figures 3 and 4 below With reference to the statistical analysis, Figure 3d shows an extremely broad standard deviation band for all ranges of cloud fraction difference. As the conclusions that follow in text lean heavily on mean

values to express model sensitivity, this is a little troubling. It is what it is, of course, but the authors should point out that the data is very noisy so that later calculations that make use of them should be taken with a grain of salt. In this case, the "very rough" results at p. 3226, line 20, should be further qualified. For example, what is the range of possible sensitivity across the distribution of O3 differences? _figure 5 below In this regard, it might be worthwhile to choose a subset of data, perhaps set of monitors in a region, and see how this cloud fraction bias works on a local or regional level.

Reply: We agree with the review's comment. We added in the text that the all data is very noisy, and analysis on the individual sites in Figure 4 can provide further information with better correlation. Correlation coefficients are also provided to show the uncertainty range. We expect that individual site's correlation can be improved if we can remove the impacts from emission (e.g. weekly pattern). Hopefully, we can pursue it in the following study.

p. 3226, line 5: -10.5ppb100%-1 looks like a typo.

Reply: Corrected

p. 3226, line 10: The use of the term "brighter" is a bit confusing here. "Brightness" is kind of a term of art in many other applications and may not be meant the same here. Is what the authors mean to say is that the model has fewer clouds?

Reply: Replaced to "have fewer clouds"

Conclusions: This is a good paper on a very interesting and timely subject. It should be published with revisions. In particular, the authors should qualify a few of their conclusions and better describe the underlying uncertainty of the data and the metrics used to estimate sensitivity, in particular the use of mean values in a noisy field of data.

Reply: Thanks again for very productive comment.

Authors' response to the review comments #2

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

Authors: Hyun Cheol Kim, Pius Lee, Fong Ngan, Youhua Tang, Hye Lim You, and Li Pan

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.

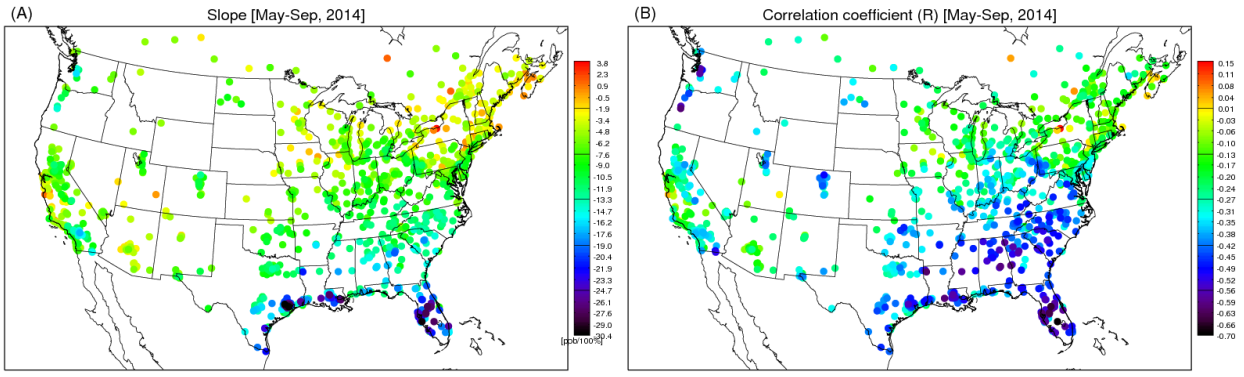

Figure R1. Spatial distributions of (a) slope and (b) correlation coefficient of linear regress between MODIS CF and MDA8 ozone.

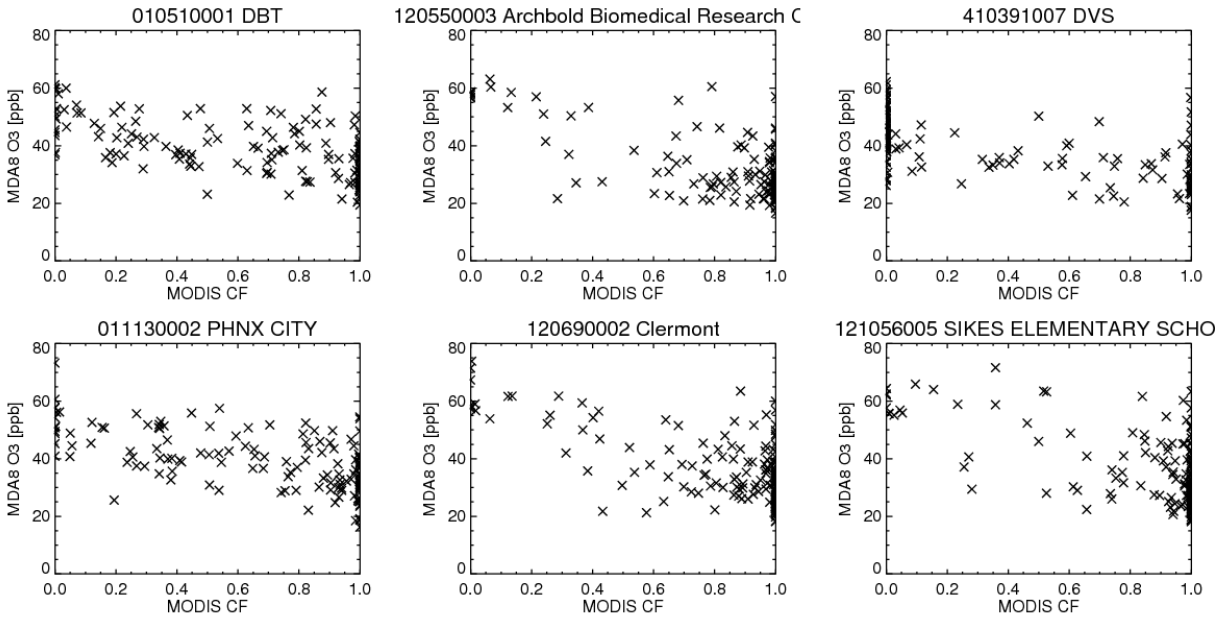

Figure R2. Scatter plots of MODIS CF & MDA8 ozone for 6 selected AQS sites.

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 relationships 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 173-184

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.

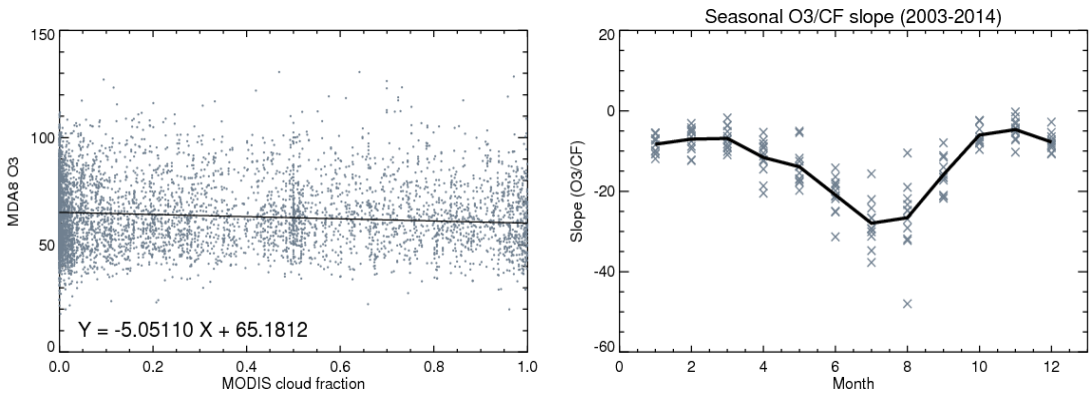

Figure R5. Scatter plot between MODIS CF and MDA8 ozone over Korea during May 20014 (left), and seasonal variations of CF-O3 sensitivity during 2003-2014 (right)

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 151-155)

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.

Evaluation of modeled surface ozone biases as a function of cloud cover fraction

Hyun Cheol Kim[1,2], Pius Lee[1], Fong Ngan[1,2], Youhua Tang[1,2], Hye Lim Yoo[1,2], and Li Pan[1,2]

[1] NOAA/Air Resources Laboratory, College Park, MD

[2] UMD/Cooperative Institute for Climate and Satellites, College Park, MD

ABSTRACT

A regional air-quality forecast system's model of surface ozone variability based on cloud coverage is evaluated using satellite-observed cloud fraction (CF) information and a surface air-quality monitoring system. We compared CF and daily maximum ozone from the National Oceanic and Atmospheric Administration's National Air Quality Forecast Capability (NOAA NAQFC) with CFs from the Moderate Resolution Imaging Spectroradiometer (MODIS) and the U.S. Environmental Protection Agency's AirNow surface ozone measurements during May to October, 2014. We found that observed surface ozone shows a ~~clear (~~negative~~)~~ correlation with the MODIS CFs, showing around 1 ppb decrease for 10% MODIS CF change over the Contiguous United States, while the correlation of modeled surface ozone with the model CFs is much weaker, showing only -0.5 ppb per 10% NAQFC CF change. Further, daytime CF differences between MODIS and NAQFC are correlated with modeled surface-ozone biases between AirNow and NAQFC, showing -1.05 ppb per 10% CF change, implying that spatial- and temporal-misplacement of the modeled cloud field might have biased modeled surface ozone-level. Current NAQFC cloud fields seem to ~~be too bright~~have less CFs compared to MODIS cloud fields (mean NAQFC CF = 0.38 and mean MODIS CF = 0.55), contributing up to 35% of surface-ozone bias in the current NAQFC system.

1. INTRODUCTION

Ground-level ozone is a secondary pollutant resulting from photochemical reactions between oxides of nitrogen (NO_x) and volatile organic compounds (VOC) in the presence of solar radiation. While local ozone production is affected by numerous factors, including precursor emissions and meteorological conditions such as temperature and local circulation, ozone photochemistry is photon-limited, and net ozone production shows a direct relationship with changes in UV actinic flux resulting from clouds and aerosols ~~(Dickerson et al., 1997; He and Carmichael, 1999; Jacobson, 1998).~~(Dickerson et al., 1997; He and Carmichael, 1999; Jacobson, 1998; Monks et al., 2004). For instance, Lefer et al. (2003) showed that without sufficient UV radiation, ozone production in Houston is limited regardless of local circulation patterns or emission sources. Studies in the urban cities of Los Angeles, California (Jacobson, 1998) and Mexico City (Castro et al., 2001; Raga, Castro et al., 2001), ~~have shown that surface ozone varies from 5% to 30% due to light-absorbing aerosols. In Houston, Lefer et al. (2003) showed that without sufficient UV radiation, ozone production is limited regardless of local circulation patterns or emission sources. Model predictions have shown an increase in the frequency of photolysis in the troposphere over the eastern United States, leading to a 5–60% increase in~~

~~lower tropospheric ozone levels due to~~also showed that surface ozone varies from 5% to 30% due to light-absorbing aerosols. Model studies have shown that surface ozone is affected by cloud fields (Voulgarakis et al., 2009; Wild et al., 2000) or strongly scattering aerosols (Dickerson et al., 1997; He and Carmichael, 1999).

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Since clouds play a critical role in the radiative balance of the Earth, their impact and models' capabilities to simulate clouds have been repeatedly tested from global and climate perspectives (Bergman and Salby, 1996; Eastman and Warren, 2013; Stephens, 2005). Clouds also play an important role in regional air quality, impacting both surface ozone and particulate matter by regulating photochemical reaction rates, heterogeneous chemistry, and the evolution and partitioning of particulate matter. These impacts, however, still have high measurement uncertainties and are not well quantified. While reliable estimates of photolysis rates are essential for reducing the uncertainty in air-quality modeling, most current models use highly parameterized methods to estimate photolysis rates. Pour-Biazar et al. (2007) argued that the uncertainties in estimation of cloud transmissivity and errors in the placement of clouds' location and time could be an important source of uncertainties in simulations of surface ozone, demonstrating during the Texas Air Quality Study campaign that surface-ozone modeling can be improved by adjusting photolysis rates based on the Geostationary Operational Environmental Satellite cloud product. They also stated that the cloud-prediction problem is particularly frustrating when modeling air quality in State Implementation Plans if they are not able to reproduce satellite-observed cloud fields in a model.

In order to reduce computational cost, most regional air-quality models, including the ~~EPA~~US Environmental Protection Agency (EPA) Community Multi-scale Air Quality model (CMAQ), use a two-step approach for calculating photolysis rates ~~(Byun and Schere, 2006).~~(Byun and Schere, 2006). In preprocessing, the clear-sky photolysis rates for a range of latitudes, altitudes, and solar zenith angles are first computed using a radiative transfer module (Madronich, 1987). Then, within the chemical-transport model, the tabular photolysis rates are interpolated for each location and then adjusted using fractional cloud-coverage information. Since most early meteorological models did not generate the full suite of specific cloud and moisture fields required as input for the chemical-transport model, regional air-quality models were designed to diagnose some additional cloud-related fields from meteorological state variables for use in the chemical-transport model. The Meteorology-Chemistry Interface Processor (MCIP), CMAQ's preprocessor, diagnoses for each horizontal grid cell the cloud coverage, cloud base and top, and the average liquid water content in the cloud using a series of simple algorithms based on a relative-humidity threshold (Otte and Pleim, 2010). For example, in CMAQ modules the photolysis rates below clouds are calculated as:

~~$$J_{below} = J_{clear}[1 + f_c(1.6 \cdot tr_c \cos(\theta) - 1)]$$~~

~~where tr_c is cloud transmissivity, f_c is the cloud fraction for a grid cell, and Θ is the solar zenith angle. Cloud fractions is estimated using relative humidity (RH) and critical RH (Geleyn et al., 1982; Schumann, 1989; Wyngaard and Brost, 1984).~~

$$J_{below} = J_{clear}[1 + f_c(1.6 \cdot tr_c \cos(\theta) - 1)] \quad (1)$$

where tr_c is cloud transmissivity, f_c is the cloud fraction for a grid cell, and Θ is the solar zenith

angle. Cloud fraction is estimated using relative humidity (RH) and critical RH (Byun and Ching, 1999). Cloud fraction (f_c^k) above the boundary layer is:

$$f_c^k = \left[\frac{RH^k - RH_c}{1 - RH_c}\right]^2 \quad (2)$$

where RH^k is the relative humidity at vertical model layer k and RH_c is the critical relative humidity defined as $RH_c = 1 - 2\sigma_c(1-\sigma_c)[1 + 1.732(\sigma_c - 0.5)]$ and $\sigma_c = p^k/p^{k_{PBL}}$ (Geleyn et al., 1982).

Within the convective boundary layer when $RH > RH_c$,

$$f_c^k = 0.34\frac{RH^k - RH_c}{1 - RH_c} \quad (3)$$

(Schumann, 1989; Wyngaard and Brost, 1984). See line 131-177 of bcldprc_ak.f90 for MCIP v3.6.

Although fractional cloud coverage (i.e., cloud fraction) thus plays a crucial role in determining the final values for photolysis rate, it is not a well-defined physical state variable and is mostly threshold-specific for each retrieval algorithm. One may notice that there are two possible uncertainties in modeling cloud fraction: (1) the model's capability to generate the proper amount of cloud fields, both in their displacement and timing; and (2) conceptual consistency in definitions of cloud fraction between model and observation (i.e., from satellite). In this study, we present efforts to evaluate the cloud-coverage information used in a regional air-quality model through satellite-based cloud fraction information and surface-monitored ozone observations. In the second section, we introduce the observational and modeling data used in this analysis, and results are discussed in Section 3. General performance of the Contiguous United States (CONUS-)-scale air-quality forecast system and possible overestimation of surface-ozone levels due to uncertainty in cloud fractions will be also discussed.

2. DATA AND METHOD

MODIS: The Moderate Resolution Imaging Spectroradiometer (MODIS) cloud level 2 product (MOD06_L2 and MYD06_L2, http://modis-atmos.gsfc.nasa.gov/MOD06_L2/index.html) is used for daily cloud-coverage information for each surface-monitoring site. We have retrieved 5-km cloud fraction data, which is based on MOD35_L2 cloud-mask information with 1km and 250m (nadir) spatial resolution. Brightness temperatures (BT) from multiple channels and their differences (BTD) are used in cloud-masking algorithms, as described in the MODIS cloud-mask product (MOD35_L2) user guide (http://modis-atmos.gsfc.nasa.gov/_docs/CMUSERSGUIDE.pdf). For example, daytime land-cloud maskings are determined using BTs and BTDs from 1.38-, 3.7-, 3.9-, 6.7-, 8-, 11-, 12-, and 13.9- µm channels. Only data from local afternoon time (~1:30 pm), when ground-level data show high ozone-production efficiency, are used in the analysis.

AirNow: Real-time ozone measurements across the CONUS are provided by the EPA through the AirNow network (http://www.epa.gov/airnow). From more than 1000 Air Quality System (AQS)

sites throughout the CONUS, hourly surface ozone data is obtained, and a daily maximum eight-hour moving averaged ozone (MDA8 ozone) value is calculated for each site.

NAQFC: The U.S. National Air Quality Forecast Capability (NAQFC) provides daily, ground-level ozone predictions using the Weather Forecasting and Research non-hydrostatic mesoscale model (WRF-NMM) and CMAQ framework across the CONUS with 12-km resolution domain (Chai et al., 2013; Eder et al., 2009). In our analysis, we used the experimental version of NAQFC, which uses WRF-NMM with B-grid (NMMB) as a meteorological driver and the CB05 chemical mechanism. Meteorological data is processed using the PREMAQ, which is a special version of MCIP designed for the NAQFC system. While NAQFC has shown a tendency to overpredict MDA8 ozone (Chai et al., 2013), recent updates to model processes and emission have reduced its bias. The "CFRAC" variable from METCRO2D output files are used for cloud fraction.

METHOD: For each EPA monitoring site and the corresponding model cells, we have calculated a daily maximum of eight-hour, forward-moving, averaged concentrations. For the same locations, we also calculated daytime (~1:30pm local time) cloud fractions from the model and from satellite data. MODIS cloud fractions are regridded into 12-km domain grid cells using a conservative regridding method (Kim et al., 2013). For consistent comparisons, only valid observational data are used, those with corresponding times and locations. We have investigated the six-month summer ozone season (May-October, 2014) and results are consistent for each month.

3. RESULTS AND DISCUSSION

General distributions of daily and monthly daytime cloud fractions from the model and from satellite are compared. Figure 1 shows the distribution of cloud fractions retrieved from NAQFC and MODIS cloud products (MOD06 level2) for one day (Aug. 2, 2014) in the upper panels; and the figure shows a one-month average (Aug. 2014) in the lower panels. The August 2 plot is overlaid with a NCEP surface-analysis chart to show its association with general features of the synoptic weather pattern. It is obvious that both model and satellite correctly display the general features of cloud coverage associated with the synoptic frontal activities. However, there is a serious discrepancy in their quantity; in most cases the amount of cloud fraction used in the model is smaller than the cloud fraction retrieved from the MODIS cloud product. For August 2014, monthly means of daytime cloud-fractions from NAQFC and MODIS are 0.38 and 0.55, respectively.

This discrepancy becomes even more evident from the histogram distribution. In Figure 2, we present histogram distributions of cloud fractions from NAQFC and from MODIS during August 2014 for each 0.1 cloud-fraction bin. Occurrence frequency is shown on the y-axis, so the sum of total frequency makes 100%. In the NAQFC model, lower cloud-fraction numbers are more dominant, with the highest frequency between 0.2 and 0.3, showing very low frequency of high cloud fractions. On the other hand, the MODIS cloud fraction is quite different, showing more of a bimodal distribution. Frequencies for clear sky are similar between the model and satellite, around 12–13%, but the satellite cloud frequency is much lower in the 0.1–0.5 range and higher above 0.6. ~~Monthly means of daytime cloud-fractions from NAQFC and MODIS are 0.38 and 0.55, respectively.~~

The reason for this discrepancy between the model and MODIS is not clear and requires future investigation. As mentioned previously, this might be a characteristic of the meteorological model or it could be a conceptual difference in cloud fraction between model and satellite. As cloud-fraction field is a diagnosed variable in PREMAQ, which uses a certain threshold of liquid-water content or relative humidity to model the existence of clouds, it may differ from the satellite's measurements of cloud, which uses emissivity-based cloud masking using BT and BTD from multiple channels.

Figures 3a and 3b show scatter plots between MODIS cloud fractions and AirNow MDA8 ozone and between NAQFC cloud fractions and MDA8 ozone, respectively, during ~~Aug.~~August 1024 across all reporting EPA AQS monitoring-sites. ~~As one readily expects from~~As the ~~basic characteristics~~amount of UVA (ultraviolet radiation in 315-399 nm) strongly affects the ozone ~~photochemistry~~production by NO_2 photodissociation (e.g. j(NO2) in $\lambda < 420$ nm) at the surface, it is evident that cloud fraction, and the eventual flux of photons reaching the level of the surface, is a very dominant component determining ground-level ozone concentration~~.~~ (Monks et al., 2004; Seinfeld and Pandis, 2006). Scatter plots in Figure 3a draw data from more than 1000 sites across the CONUS under a variety of meteorological conditions and precursor sources. Even with the high uncertainties here, we can see a ~~clear~~notable separation of ground-level ozone for each cloud-fraction bin, implying that photon flux is one of the most dominant features determining tropospheric ozone photochemistry. Slope and offsets for line-fitting MODIS CF versus AirNow MDA8 ozone are -11.~~39~~33 and 49, respectively, implying that 10% of CF change can cause around 1.~~14~~13 ppb decrease in surface ozone. On the other hand, the correlation between NAQFC CF and MDA8 ozone is slightly weaker (Figure 3b); slope and offsets between NAQFC CF and MDA 8 ozone are -5.0 and 50.5, respectively, showing half as much sensitivity in surface ozone according to the NAQFC CF compared to the MODIS CF.

Figures 3c and 3d are scatter plots for CF differences (NAQFC-MODIS) and MDA8 surface ozone bias (NAQFC-AQS; left), and averaged O_3 biases for each 0.1 cloud-fraction bin (right). Since the definition of cloud fraction in the model and the satellite are slightly different, we choose the term "cloud fraction difference" instead of "cloud fraction bias." ~~Correlation slope~~Slope of the linear regression is -10.5 ppb/100% CF. The right-side panel shows averages of ozone biases for each 0.1 bin. The vertical bars indicate 1 standard deviation. It is clear that where the model underestimates cloud fraction, it likely overestimates surface ozone, although there are many intricacies of tropospheric ozone chemistry involved.

Since Figure 3 shows data from all AQS sites, it includes multiple uncertainties from each site's local characteristics, such as local emissions. We have conducted further investigation for individual AQS sites to confirm if we can find similar MDA8 ozone to CF correlation. Figure 4 shows spatial distributions of each site's ozone to CF sensitivity (e.g., regression slope of MDA8 ozone and CF) and correlation coefficients during 5 months (May to September, 2014). MDA8 ozone decreases rapidly by the increase of CF in the southern regions, especially near the coastal lines of Gulf of Mexico, such as Texas, Louisiana and Florida, up to -30 ppb/CF. In the middle latitude regression slopes are around -10 ppb/CF, and some northern location areas show positive correlation. Mean of total regression slope is -8.5 ppb/CF. Correlation coefficients (R) also show stronger (negative) correlation in southern states, especially southeastern US up to R=-0.7 while northeastern US shows much weaker correlation, implying accurate CF information is important in southern US states.

OZONE OVERPREDICTION : As already described, current NAQFC cloud fields seem to ~~be brighter~~ have fewer clouds than MODIS ~~cloud fields~~ by 0.2. We have further estimated how this difference can affect the general performance of surface ozone forecast. Previous studies address O_3 overpredictions of global and regional chemical-transport models during the summer daytime over the eastern United States (Chai et al., 2013; Eder et al., 2009; Fiore et al., 2009; Murazaki and Hess, 2006; Nolte et al., 2008; Rasmussen et al., 2012; Reidmiller et al., 2009). Studies have addressed that the vertical resolution (Murazaki and Hess, 2006), the coarse representation of emissions (Liang and Jacobson, 2000), along with uncertainty in the heterogeneous reactions of aerosols (Martin et al., 2003) contribute to the highly biased O_3 of the global chemical-transport models MOZART or GEOS-Chem over the eastern United States. NAQFC also has a tendency to overestimate surface ozone during ozone season. We may estimate the amount of possible overestimation of surface ozone due to the underestimation of the cloud fraction and eventual overestimation of photolysis rate. As the mean cloud fraction of model is 0.17 higher than the cloud-fraction estimated from MODIS, by applying the -10.5 ppb/CF estimate, we can deduce that 1.8 ppb of the surface-ozone overestimation is contributed from the underestimation of the cloud fraction. Considering current NAQFC surface-ozone overestimation is around 5 ppb for the month of August 2014, we can roughly suggest that almost 35% of this overestimation is due to faulty estimation of the cloud field. Though this estimate is still very rough, this is definitely something to consider carefully in order to improve the simulation of regional air quality and especially the simulation of surface ozone.

RESOLUTION ISSUE: In utilizing satellite-based cloud-fraction information, one concern is how to process data in terms of pixel resolution. As already mentioned, the cloud fraction is not a state variable; it is threshold- or retrieval-specific. For example, if we consider an area with 9 pixels with cloud fraction 0.6, fractional averaging of 9 cloud pixels should yield a 0.6 cloud fraction. However, if we first perform cloud masking for each pixel, we may have 9 cloud markings out of 9 pixels, resulting in 100% cloud fraction. This might not be a critical error on a global scale, but it is a crucial difference for regional or local scales intended for investigating the spatial scale of local ozone production. Since cloud fields are very localized phenomena, this information should be processed as finely as data are available.

To conclude, this study demonstrates that appropriate model of CF is crucial in the modeling of surface ozone chemistry. Further studies are needed in terms of the comparison of modeled- or satellite-based CF with actual surface level photon flux, as well as enhanced parameterization of CF in the air quality model.

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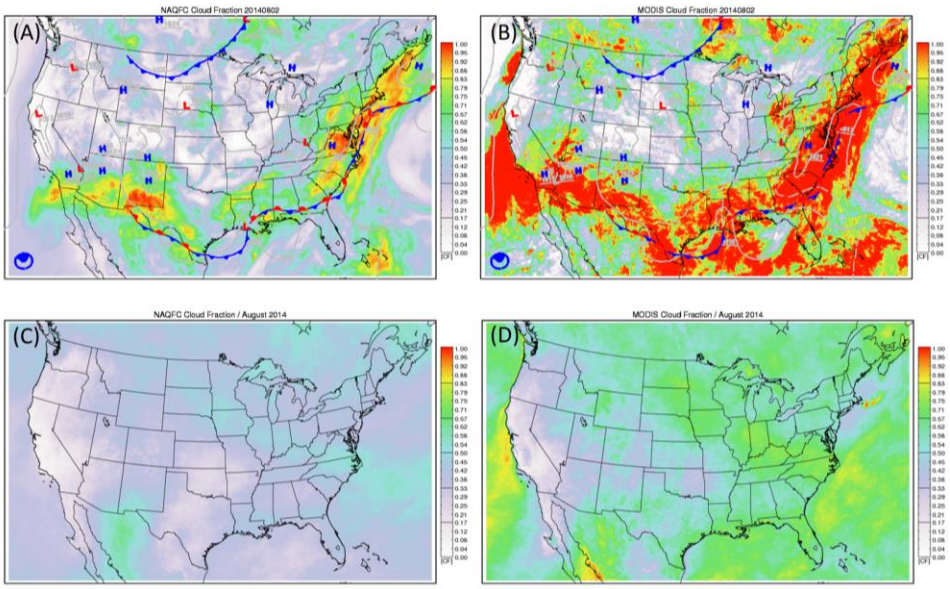
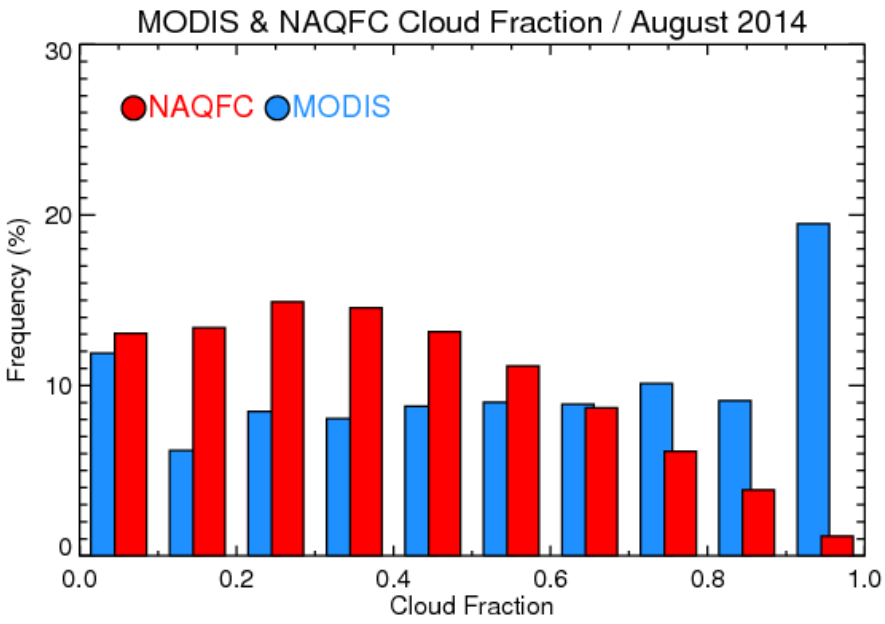


Figure 1. Spatial distributions of cloud fractions on Aug. 2, 2014 from NAQFC (a) and MODIS (b). NOAA NCEP surface weather chart at 18UTC is overlaid. Monthly averaged distributions are also shown for NAQFC (c) and MODIS (d).



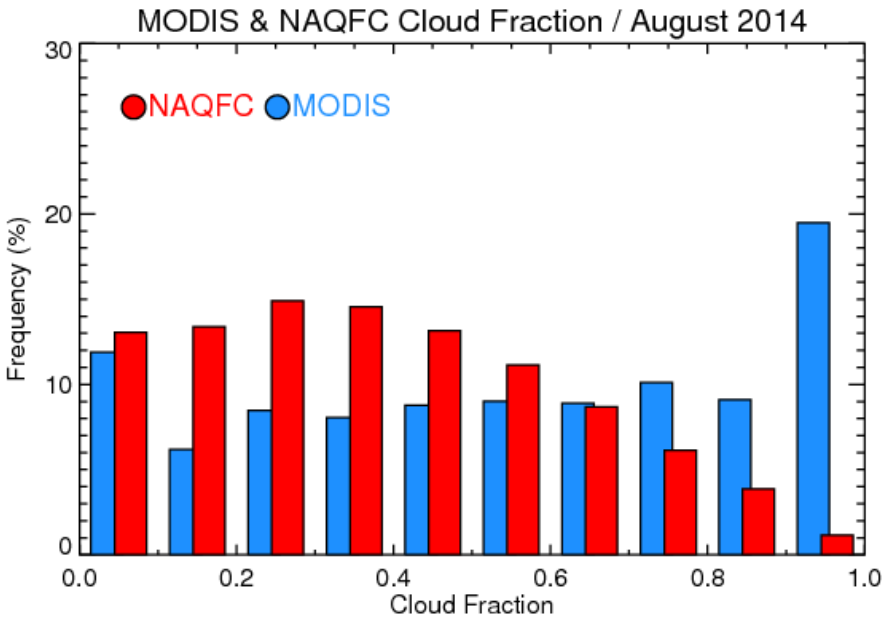


Figure 2. Occurrence frequency histogram for NAQFC cloud fractions (red) and MODIS cloud fractions (blue).

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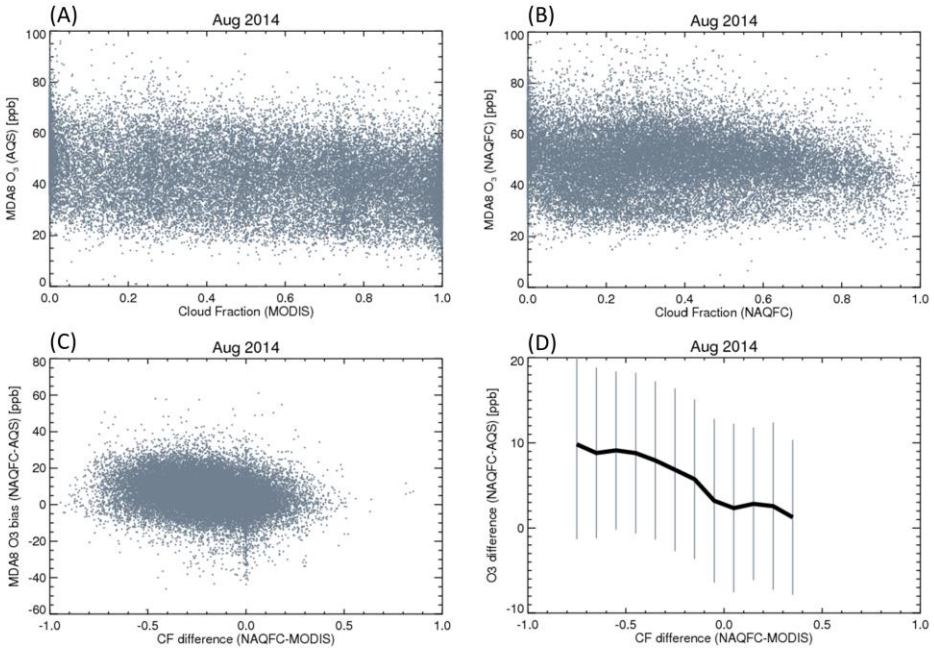

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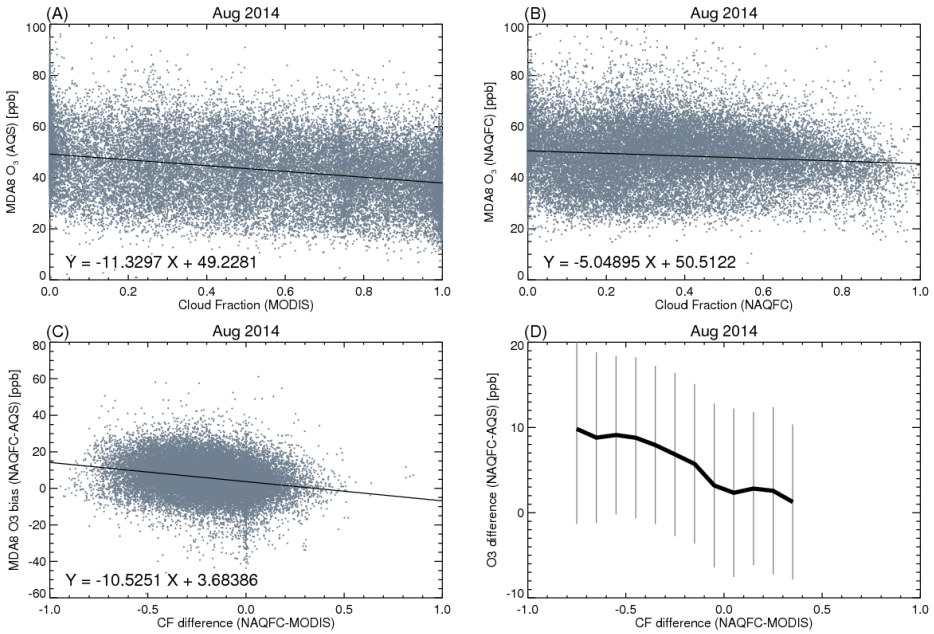

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Figure 3. Scattered plots between MODIS cloud fractions and AQS MDA8 ozone (a), between NAQFC cloud fractions and MDA8 ozone (b), and between cloud fraction differences (NAQFC - MODIS) and MDA8 surface ozone bias (NAQFC-AQS) (c) during Aug. 2014 across 1024 AQS monitoring site locations. Averaged O_3 biases for each 0.1 cloud-fraction bin with 1 standard deviation (vertical bars) are also shown (d).

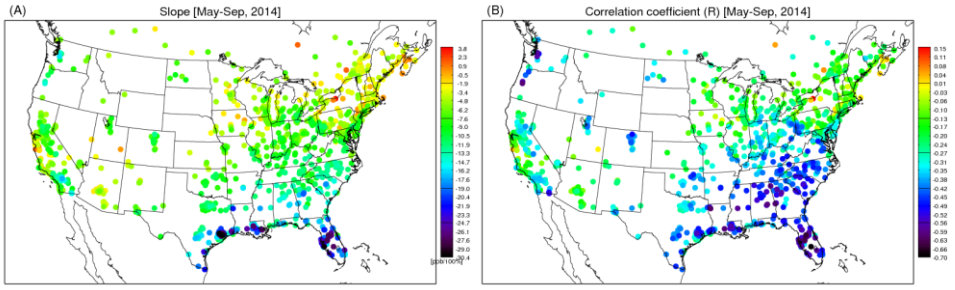

Figure 4. Spatial distributions of (a) slope and (b) correlation coefficient of linear regression between MODIS CF and MDA8 ozone.