Authors' Responses (in red) to the Comments on acp-2019-642 by Anonymous Referee # 1 and Changes Made (in blue) in the Manuscript

This manuscript separated short-term synoptic-scale fluctuations from long-term baseline component embedded in the daily maximum 8-hr ozone time series using a filter and estimated the limit of air quality model's accuracy (or predictability/uncertainties of air quality prediction). This is an interesting topic for air quality prediction.

The authors thank the referee for recognizing the importance of our work.

But to my surprise, the authors did not even consider lead time when discussing air quality predictability (or limit of air quality prediction). What is the configuration of the air quality prediction? Was this one-day prediction? Two-day prediction? Prediction uncertainties/errors will change significantly with different lead time.

It seems that the referee has misunderstood the modeling simulations we have examined in this paper. Please note that our paper focused on the evaluation of retrospective simulations of 21-years (1990 to 2000) of the daily maximum 8-hour average ozone concentrations over the contiguous United States (CONUS) with the fully-coupled Weather Research Forecasting (WRF) meteorological model and the Community Multiscale Air Quality (CMAQ) chemical transport model. While the concepts presented in our paper are applicable to examining air quality forecasting products, the results of our study reflect the prediction capability of the model based on retrospective simulations and **not** air quality forecasting. To ensure better characterization of the prevailing meteorology (i.e., synoptic forcing) for these retrospective 21-year simulations, four-dimensional data assimilation (FDDA) was utilized following the methodology suggested by Gilliam et al. (see *Atmos. Environ.*, Vol.53, pp 186-201, 2012) and modified for fully-coupled meteorology-chemistry model applications as described in Hogrefe et al. (see *Atmos. Environ.*, Vol. 115, pp 683-694, 2015). The modeling set-up and performance evaluation of these historical multiyear WRF-CMAQ simulations have been published by Xing et al. (2015), Gan et al. (2015), and Astitha et al. (2017) as referenced in our manuscript. The following material has been added in the revised manuscript.

Expanded the model description in Section 2 (added after page 3 line 7 in the original manuscript) as follows: "To ensure better characterization of the prevailing meteorology (i.e., synoptic forcing) in the retrospective 21-year WRF-CMAQ simulations, four-dimensional data assimilation (FDDA) was utilized following the methodology suggested by Gilliam et al. (2012) and modified for fully-coupled meteorology-chemistry model applications as described in Hogrefe et al. (2015). The model set-up and performance evaluation of these historical multiyear WRF-CMAQ simulations have been published by Xing et al. (2015), Gan et al. (2015), and Astitha et al. (2017)."

It is also surprising to see the authors suggesting improving simulation of the baseline concentration by focusing on the quality of the emission inventory and the model's treatment for the slow-changing atmospheric processes. I have no question for improving emission inventory, but I am confused by improving the slow-changing atmospheric processes.

A number of papers have been published, documenting the importance of the baseline (longer-term) forcing embedded in ambient ozone data (see e.g., Rao et al., 1996 and 1997; Hogrefe et al., 2000; Rao et al., 2011; Porter et al., 2017; Astitha et al., 2017; Luo et al., 2019). As noted in Astitha et al. (2017), the baseline level and the strength of the synoptic forcing are to be viewed as the necessary and sufficient conditions for observing peak ozone levels. Rao et al. (2011), Hogrefe et al. (2000), Astitha et al. (2017), Porter et al (2017), and Luo et al. (2019) have demonstrated that when the magnitude of the baseline level is low, there will not be ozone exceedances of the USA's National Ambient Air Quality Standard no matter how strong the synoptic forcing is. In this study, we are working with the model (WRF-CMAQ)-predicted and corresponding observed time series of the daily maximum 8-hour ozone concentrations during 1990 to 2000 at various monitoring locations over CONUS; therefore, the Nyquist interval here is 2-days. Using both Empirical Mode Decomposition (EMD) and KZ filtering, we separated the synoptic forcing (time scale < 24 days) and baseline (time scale > 1 month) forcing embedded in the time series of observed and modeled daily maximum 8-hour ozone concentrations. To illustrate, the results of the application of EMD to the daily maximum 8-hr ozone time series data measured at Altoona, PA are presented in Fig. 1 below. The top left panel displays the raw ozone time series while the top of the right panel shows its power spectrum. The 7 intrinsic mode functions (IMFs) and the residual on the left side, and their corresponding power spectra on the right reveal that most of the synoptic-scale features in ozone data are reflected in IMFs 1 and 2. The baseline ozone is extracted by removing the first two IMFs from the raw ozone time series.

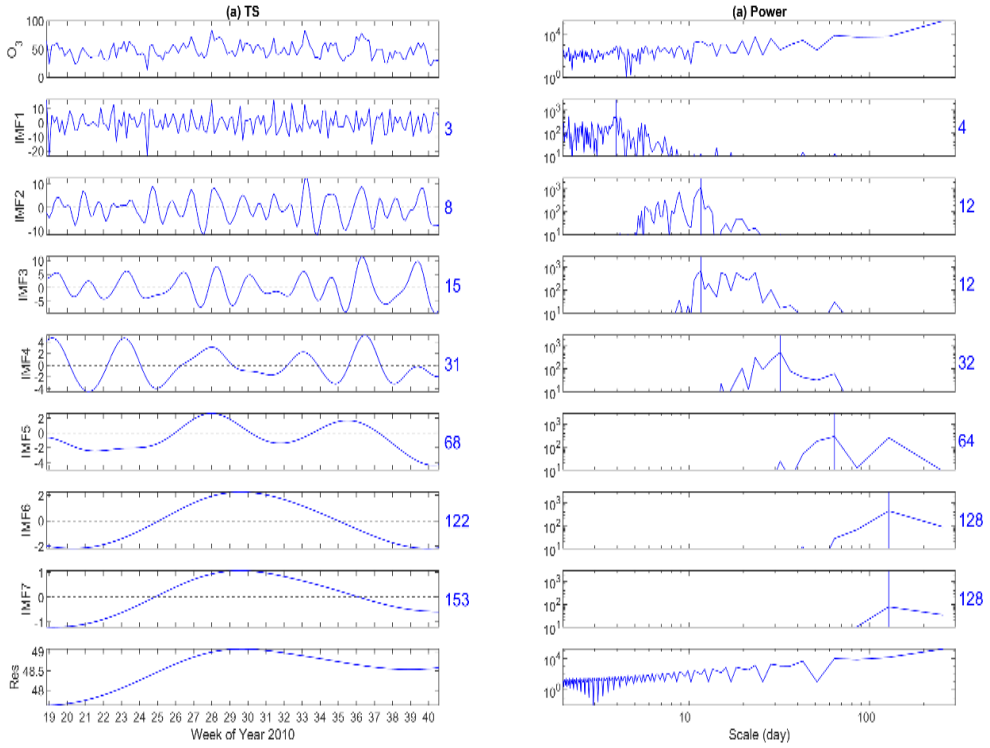


Figure 1. Results of the application of the EMD technique, which is designed for analyzing non-stationary and non-linear time series data, to the daily maximum 8-hour ozone time series data at the Altoona, PA site. The numbers on the right side represent the time scale (in days) associated with each IMF. Note, the power spectrum of raw ozone time series shows that the energy in the 1-10 days (SY forcing) is an order of magnitude less than that in the longer (baseline) time scale.

The scale separation achieved from the application of EMD and KZ filter, displayed in Fig. 2, reveal that the results are quite similar. It should be noted that the short-term component (SY) extracted from the KZ filter as well as EMD's high-frequency IMFs 1 and 2 resemble white noise process.

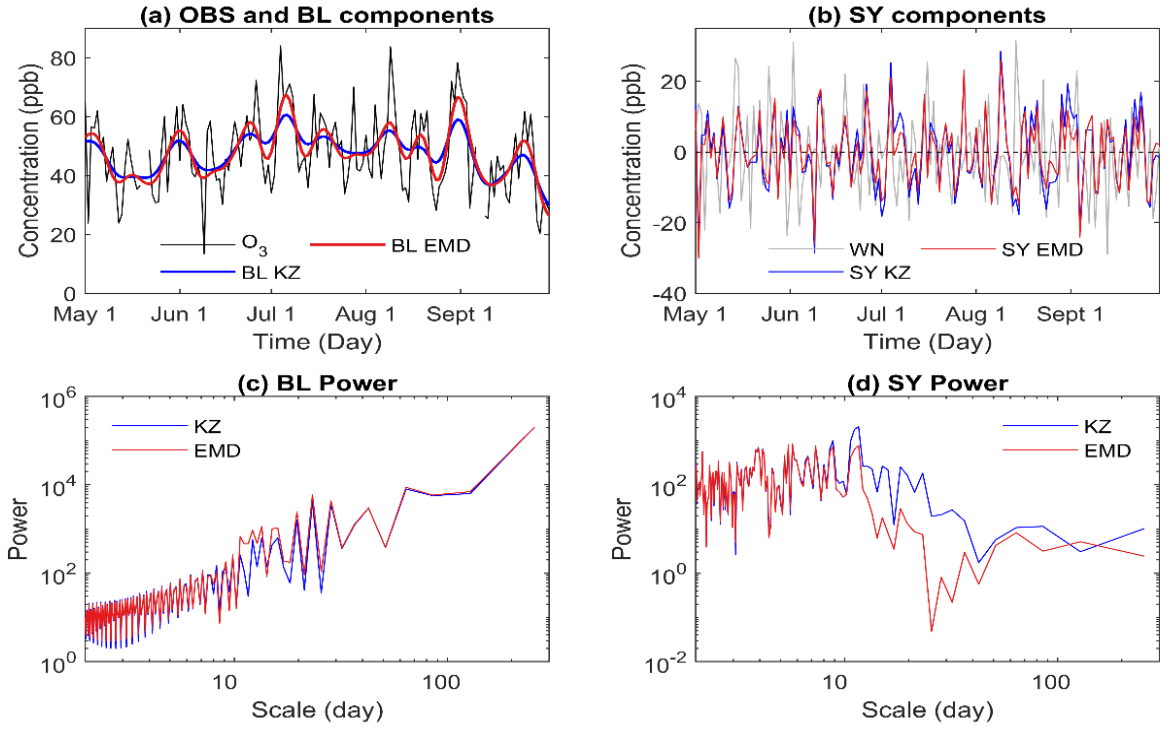


Figure 2. The top left panel displays the raw daily maximum 8-hour ozone time series together with the baselines extracted from the KZ filter and EMD while the top right panel reveals the similarity between white noise and synoptic forcing imbedded in these observed ozone time series. The bottom two panels compare the power spectra of the baseline forcing (left) and the synoptic forcing (right) derived from KZ filtering and EMD (sum of IMF 1 and IMF2). Notice that most of the energy in the baseline time series is in the longer time scale while most of the energy of the short-term component is in the high-frequency range. The similarity of results from both scale separation techniques demonstrates that the two scales of interest (i.e., baseline and synoptic forcing) have been extracted reasonably well.

This material to demonstrate good scale separation has been added in the revised manuscript.

Added new material to the beginning of Section 3.1 (page 4 line 15 in the original manuscript): "Using both Improved CEEMDAN and KZ filtering, we separated the synoptic forcing (time scale < 24 days) and baseline (time scale > 1 month) forcing embedded in the time series of observed and modeled daily maximum 8-hour ozone concentrations. To illustrate, the results from the application of Improved CEEMDAN to the daily maximum 8-hr ozone time series data measured at Altoona, PA are presented in

Fig. 1. The top left panel displays the raw ozone time series while the top of the right panel shows its power spectrum. The 7 intrinsic mode functions (IMFs) and the residual on the left side, and their corresponding power spectra on the right reveal that most of the synoptic-scale features in ozone data are imbedded in IMFs 1 and 2. The baseline ozone is extracted by removing the first two IMFs from the raw ozone time series. To illustrate the concept of the ozone baseline, DM8HR time series measured in 2010 at Altoona, PA is presented in Fig. 2a together with the embedded baseline concentration as extracted by the KZ5,5 and Improved CEEMDAN. It is evident that high ozone levels are always associated with the elevated baseline. The difference between the raw ozone time series and baseline, denoted as the short-term or synoptic forcing (SY), is displayed in Fig. 2b. The power spectra, displayed in Figs. 2c and d, reveal both methods yielded good scale separation. Due to the good agreement between both scale separation techniques, only the results from the KZ filter are presented for the remainder of the manuscript."

The huge advances of weather prediction during the past few decades has been focusing on 1-day or 2-day prediction. On such short-term synoptic-scale weather processes, our weather prediction did excellent job and has been improved through years. Such improvement can benefit air quality prediction significantly (Zhang et al., 2007). I would thus imagine that such short-term practical predictability of air quality can be much improved through better model treatments and better initial conditions of meteorological and chemical variables, as well as emissions.

As stated before, our study deals with evaluating the retrospective air quality simulations, not modeling results from an air quality forecasting effort. Air quality modeling uncertainty even for the retrospective modeling cases, outside of the chemistry formulation and boundary conditions, is attributed primarily to meteorology and emissions inputs. Vautard et al. (*Atm. Environ*., Vol. 53, pp 15-37, 2012) concluded that major challenges still remain in the simulation of prevailing meteorology (e.g., errors in wind speed, PBL, nighttime meteorology, clouds) in retrospective air quality modeling. Based on retrospective ozone episodic modeling with the WRF-CMAQ model using various sets of equally likely initial conditions for meteorology along with FDDA, Gilliam et al. (2015) confirmed the presence of sizable spread in WRF solutions, including common weather variables of temperature, wind, boundary layer depth, clouds, and radiation, thereby causing a relatively large range of ozone concentrations. Also, pollutant transport is altered by hundreds of kilometers over several days. Ozone concentrations of the ensemble varied as much as 10–20 ppb (or 20–30%) in areas that typically have higher pollution levels.

We acknowledge that air quality forecasting often is concerned with accurately predicting transient pollution events. The analysis of modeling uncertainty in our retrospective simulations that employed data assimilation suggest that the largest improvement in forecast accuracy can be achieved through implementing bias correction techniques (i.e. by correcting for systematic errors such as those caused by emission uncertainties that manifest themselves primarily in the baseline) as demonstrated by Kang et al. (2008 *JGR-Atmospheres*, Vol. 113, Issue D23308; 2010 *Atmos. Environ*., Vol. 44, 18, pp 2203-2212). However, we do not dispute the fact that any advances in predicting short-term atmospheric processes or phenomena, be it through model improvements, better initial conditions, or ensemble techniques, may help to reduce the portion of the prediction error that is not due to systematic biases. Rather, we argue through our analysis of both observations and retrospective air quality simulations that, at least

for retrospective air quality planning applications, the focus of model development and evaluation efforts should be on longer time scales. The following discussion has been included in the revised manuscript.

Expanded the relevant sentences in the conclusions (page 7, lines 9 – 11 of the original manuscript) as follows: "To improve regional-scale ozone air quality models, attention should be paid to accurately simulate the baseline concentration by focusing on the quality of the emission inventory and the model's treatment for the boundary conditions and slow-changing (operating on sub-seasonal, seasonal, and longer-term time scales) atmospheric processes. Also, errors in reproducing the synoptic forcing can possibly be reduced with high-resolution meteorological modeling using appropriate data assimilation techniques."

Other specific comments: Many of the concept/discussion regarding inherent/practical predictability, reducible/irreducible uncertainties are questionable/wrong, or different from those used in weather prediction. For example, emissions are definitely reducible uncertainties and factors in practical predictability. Please carefully define those terms/concepts and refer to normally used/accepted definitions. The writing is overly concise, particularly in many cases where detailed explanation is needed. References: Zhang, F., Bei, N., Nielsen-Gammon, J. W., Li, G., Zhang, R., Stuart, A., & Aksoy, A. (2007). Impacts of meteorological uncertainties on ozone pollution predictability estimated through meteorological and photochemical ensemble forecasts. Journal of Geophysical Research, 112, D04304. https://doi.org/10.1029/2006JD007429 Interactive comment on Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2019-642, 2019.

It is difficult to identify exactly which terms the reviewer is referring to in comment that terms are not defined or are not defined as "normally used/accepted". From our perspective, we are consistent with the reviewer's example (we agree that uncertainty in retrospectively simulated ozone concentrations can be improved with improvement in emissions characterizations). The confusion in the definition of terms may be more to do with the reviewer's misunderstanding that the manuscript was focused on air quality forecasting rather than on analysis of retrospective simulations. We agree that terminologies between the weather forecasting community and the air quality modeling community may differ, but we believe that most air quality modelers are familiar with the terminology used in this paper. We respectfully request the reviewer to read the articles by Rao et al. (January 2011 issue of the *Bull. Amer. Meteor. Soc.*, pp 23-30), Dennis et al. (2010), Solazzo and Galmarini (2015), and Gilliam et al. (2015) included in the reference list.

Authors' Response (in red) to Referee #2's Comments and Changes in Manuscript (in blue)

The paper presents some interesting findings about inherent uncertainties in chemistry transport model simulations of ozone concentrations. It is very well written, but sometimes hard to follow. In my opinion, some terms should be explained in more detail, before the paper can be published.

We thank the reviewer for the positive feedback on our paper.

General comments:

The authors should explain why they think the data set they constructed by combining measured base line ozone with meteorology related short term variations from a 21 years CMAQ run could be seen as the output of a "perfect model". They claim that there is some inherent variability in the meteorological data that cannot be captured by any model system. However, reanalysis data may represent meteorologically related variations on time scales of few days very well, i.e. part of the variation included in the SY component of the time series may be modelled quite well.

We have revised the discussion in Section 3.2 to clarify that this analysis combining the observed baseline component with 21 CMAQ synoptic components is meant to quantify the amount of model error present in the current simulations that could conceivably be reduced through improving the representation of synoptic-scale processes and/or increased horizontal resolution. As part of this revision, we no longer refer to this combination of the measured baseline and modeled synoptic component as "perfect model". Moreover, we have added text at the end of the introduction to clarify that when we refer to the errors that can be expected even from a "perfect" model with "perfect" inputs throughout the manuscript, we consider these errors to be those arising from atmospheric stochasticity which we estimate in Section 3.1 using historic observations. We also should have noted in our original manuscript that four-dimensional data assimilation (FDDA) was utilized following the methodology suggested by Gilliam et al. (see *Atmos. Environ*., Vol.53, pp 186-201, 2012) and modified for fully-coupled meteorology-chemistry model applications as described in Hogrefe et al. (see *Atmos. Environ.*, Vol. 115, pp 683-694, 2015). As the reviewer pointed out, the SY component in the model output contains some meteorologically related variations on time scales of few days. The following revisions and additions have been made to address the reviewer's comment:

Revised and expanded the end of Section 1 (page 2, lines 29 – 32 in the original manuscript) as follows: "Also, no assessments have been made to date on the errors that are to be expected even from "perfect" regional-scale air quality modeling systems. To estimate such irreducible model errors due to atmospheric stochasticity (which we consider to be the errors that are expected even from a "perfect" model with "perfect" inputs), we analyzed the observed daily maximum 8-hr (DM8HR) ozone time series data at monitoring locations across the contiguous United States (CONUS) during the 1981-2014 time period and present the results of this analysis in Section 3.1. In Section 3.2, we illustrate how this information could be used in guiding model development specifically aimed at addressing reducible errors in the synoptic component by contrasting the results from Section 3.1 with analysis using the

synoptic component from a 21-year simulation performed with the fully coupled WRF-CMAQ simulations covering the 1990-2010 period."

Expanded the model description in Section 2 (added after page 3 line 7 in the original manuscript) as follows: "To ensure better characterization of the prevailing meteorology (i.e., synoptic forcing) in the retrospective 21-year WRF-CMAQ simulations, four-dimensional data assimilation (FDDA) was utilized following the methodology suggested by Gilliam et al. (2012) and modified for fully-coupled meteorology-chemistry model applications as described in Hogrefe et al. (2015). The model set-up and performance evaluation of these historical multiyear WRF-CMAQ simulations have been published by Xing et al. (2015), Gan et al. (2015), and Astitha et al. (2017)."

Modified the discussion in Section 3.2 (page 5, line 23 – page 6, line 9 in the original manuscript) as follows: "In this section, we analyze long-term records of model simulations in an attempt to quantify the error associated with the modeled SY component that results both from not explicitly representing stochastic variations in atmospheric dynamics and emission variability in the current generation regional air quality models and from other reducible sources of model error. … To isolate the impact of model imperfections on only the SY time scale on errors across the ozone distribution, we assume that the model perfectly reproduces the 'true' BL depicted by the observed 2010 BL. We then use this 'perfect' modeled BL and reconstruct 'pseudo-simulated' ozone time series, similar to what was done in Fig. 3, except for using the SY component embedded in the 21 years of coupled WRF-CMAQ simulations. The rationale for this analysis is to quantify the amount of model error present in the current simulations that could conceivably be reduced through improving the representation of synoptic and mesoscale processes and/or increased horizontal resolution with appropriate data assimilation techniques."

Why do you use 30+ years of ozone measurements for analyzing the observations while only 21 years can be used for comparisons to the model results? Wouldn't it be enough to look at the data set 1990 to 2010 for the observations, too? And why do you need to construct "pseudo ozone observations" and cannot use the observational data set as such? Please explain this in the text.

As noted in our paper, any observation at a given percentile represents an event or a single realization out of a population. The object of our paper is to quantify the inherent variability in the observations due to the stochastic nature of the atmosphere. To this end, we thought that the use of 30+ years of historical data rather than 21 years would help in making more robust estimates of the expected errors even from "perfect" models driven with "perfect" input. As demonstrated in our previous research (see Porter, et al., 2017 *Atm. Poll. Res*.; Astitha, et al., 2017 and Luo, et al., 2019 in *Atm. Env*.), the baseline forcing can be viewed as the deterministic part in observations while the SY forcing is the near-stochastic part. We superimposed 30+ adjusted SY forcing on the baseline embedded in 2010 ozone observations to generate 30+ representations of ozone concentrations, which are equally likely to occur at any given probability point stemming from the stochastic nature of the atmosphere. Details on this approach can be found in Luo, et al. (2019 AE). The discussion in Section 3.1 of the revised manuscript was modified in response to the reviewer's comments.

Updated the second paragraph of Section 3.1 (page 4, line 25 – page 5, line 11 in the original manuscript) as follows: "Once the scale separation is achieved with the KZ5,5, we superimposed the SY forcing imbedded in 30+ years of historical DM8HR ozone time series measured at a given location on the baseline component of the ozone time series at that location to generate 30+ reconstructed or pseudo ozone distributions. Illustrative results using eq. (3) at a suburban location in Altoona, PA are presented for 2010 base year in Fig. 3a … note, it is equally likely for any of these 30+ CDFs to occur because of the stochastic nature of the atmosphere even though the individual event in 2010 yielded the CDF shown in red. As mentioned before, ozone mixing ratio at any given probability point on the red line in Fig. 3a reflects an individual event while ozone values at the same probability in different CDFs (gray lines) reflect the population stemming from the stochastic nature of the atmosphere. In other words, there are 30+ dynamically consistent ozone time series attributable to the 2010 baseline (given 2010 emissions) for examining the inherent variability due to atmospheric stochasticity. … Using these 30+ pseudo-observation ozone mixing ratios and the actual observed ozone values at each percentile, statistical metrics such as Bias, RMSE, coefficient of variation (CV=standard deviation/mean), normalized mean error (NME) and normalized mean bias (NMB) are presented in Fig. 3b and c (see Emery et al. (2016) for the description of the statistical metrics considered here). … The extreme values are better described in statistical terms rather than in deterministic sense (Hogrefe and Rao, 2001)."

Specific comments:

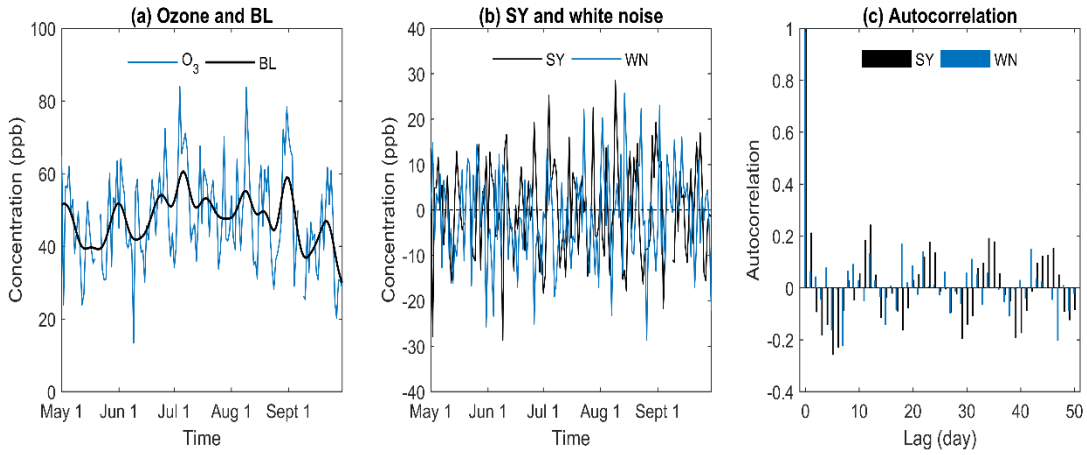
Page 3, line 26: In equation (1) it should be made clear that the filter KZ(5,5) is applied to the ozone time series O3(t).

Yes, the reviewer is correct that the SY component is estimated by applying the KZ filter with a window length of 5 days and 5 iterations to the ozone time series O3(t) as described in Porter et al. (2015), Rao et al. (2011), and Luo et al. (2019). We updated the notation in equations (1) and (2) and also changed occurrences of KZ(5,5) in the text to $KZ_{5,5}$ to better reflect the operation of the filter.

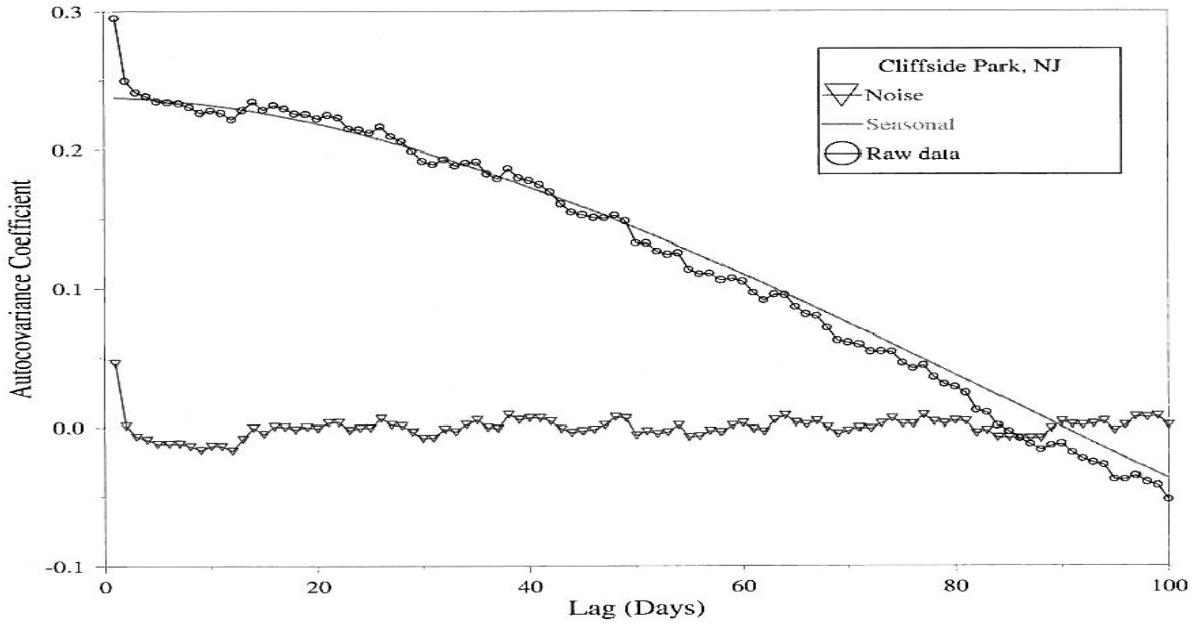
Equations 1 and 2: changed "KZ(5,5)" to "$KZ_{5,5}(O_3(t))$". Also changed all occurrences of KZ(5,5) in the text to $KZ_{5,5}$

Page 4, line 17: Can you show by statistical evaluation that the SY component represents white noise.

Because we haven't used the concept of white noise process to statistically model the SY forcing in our paper, we've removed the reference to white noise in the revised manuscript. However, to respond to the referee's question, we display below the autocorrelation function for time series in SY forcing and white noise. It is evident that the correlation drops off after 1-day lag.



In addition, we display below the autocovariance function as a function of lag (days), extracted from the article titled "Dealing with the ozone non-attainment problem in the Eastern United States" by Rao et al. on Page 20 in the January 1996 issue of the EM Magazine, a publication of the Air & Waste Management Association. These results reveal that the short-term variation (SY component) in observed ozone time series data is statistically indistinguishable from "white noise" with an autocorrelation coefficient that drops to zero at a lag of 1 day.



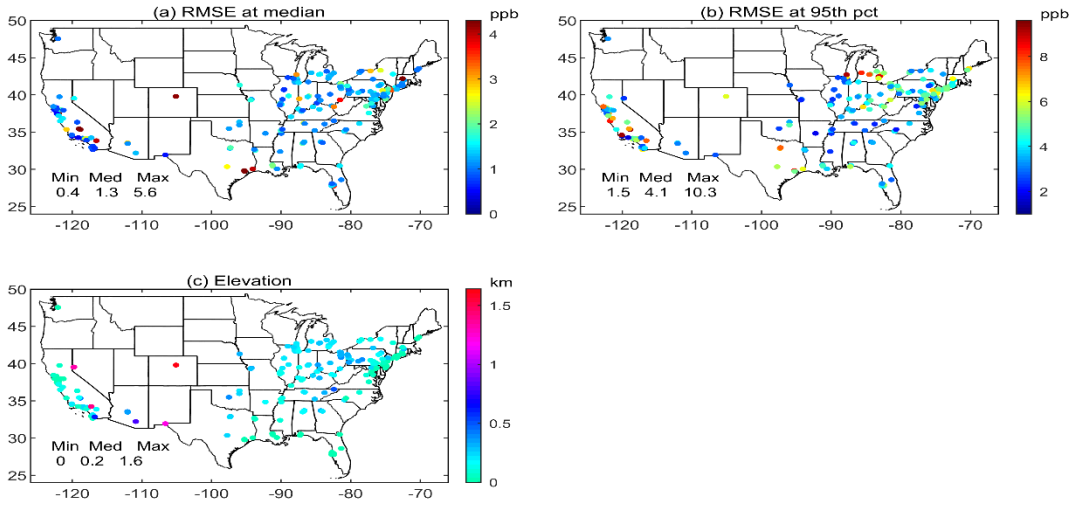
Page 4, line 18: please explain AR(1)

An AR(1) autoregressive process is the first-order process, meaning that the current value is based on the immediately preceding value. However, since we removed the references to "white noise" and AR(1) from our manuscript, this explanation has not been added to the revised manuscript.

Page 5, line 18-20: It isn't obvious for the reader which stations are at elevated sites.

We agree that adding this information would be useful for the reader and have added a panel to Figure 5 that shows the elevation of each monitoring site over CONUS.

The following new figure (Figure 5c) has been added to the revised manuscript:



Page 5, line 25: Define the "strength" of the SY component (being the standard deviation of the time series) here.

The definition has been added to the revised manuscript as follows:

"it should be noted that the linear relationship between the strength of SY (defined as the standard deviation of the data in the synoptic component) and the magnitude of the BL (defined as the mean of the data in the baseline component) has been taken into account in generating 30+ years of adjusted SY forcing as illustrated in Luo et al. (2019)"

Page 6, line 8: Is there any reason why you selected this site?

It has complete data for 30+ years. We could have picked another site; they all exhibit same features.

Page 6, line 10/11: Is there an explanation why the model doesn't perform well for low concentrations?

The CMAQ team at the U.S. Environmental Protection Agency is investigating the reasons for model's poor performance at the low end of the concentration distribution. Plausible causes include 36-km grid spacing not resolving the effects of NO titration in urban areas, errors in atmospheric deposition, representation of variability in background concentrations of O_3, precursor and reservoir species, etc. Also, it should be noted that the stochastic variability affects mostly the lower and upper tails of the pollutant concentration distribution.

Page 6, line 31/32: Couldn't this also be caused by emissions missing the correct temporal variation?

Yes, it is possible. To address the reviewer's comment, we have expanded an earlier sentence that also discusses the limitations of the current model setup as follows:

Revised sentence (page 6, lines 1-2 in the original manuscript): "The 36-km grid may be better representing the large-scale synoptic forcing associated with the translation of weather systems than the meso-scale weather and urban influences (both dynamics and emission driven) that are embedded in the observed SY component."

Page 7, line 9: How could these "slow changing processes" be improved in the models? What is the role of stratosphere/troposphere exchange which – to my knowledge – isn't well represented in the CMAQ model runs.

As already indicated in the manuscript discussion, one needs to pay more attention to properly specifying the lateral boundary conditions, duration/strength of stratosphere-troposphere exchanges, Madden-Julian Oscillation (MJO), ENSO, climate change, control policies, spatio-temporal variability in emissions loading, etc. The hemispheric CMAQ simulations used to drive the regional CMAQ runs used here employed potential vorticity-based scaling to represent O_3 in the model's upper troposphere-lower stratosphere (UTLS). The method was subsequently enhanced to represent seasonal and latitudinal dependencies in the relationship between potential vorticity and ozone and improved the 3-dimensional O_3 distribution represented by the model as well as the seasonal impacts of STE on lower tropospheric and surface-level O_3 as detailed in analyses presented in Xing et al. (2016) and Mathur et al. (2017). Interested readers are pointed to these studies and references are also included in the discussion.

Modified the second paragraph of Section 2 (Page 3, Lines 11-19 in the original manuscript) and added two references as follows: "It has been shown that time series of the daily maximum 8-hr ozone concentrations contain fluctuations operating on different time scales (e.g., intra-day forcing induced by the fast-changing emissions and atmospheric boundary layer evolution; diurnal forcing induced by the day and night differences; synoptic forcing induced by the passage of weather systems across the country, sub-seasonal forcing due to Madden-Julian Oscillation (MJO), and long-term forcing induced by emissions, El-Nino-Southern Oscillation (ENSO), climate change, and other slow-varying processes such as seasonal and sub-seasonal variations in the atmospheric deposition and stratosphere-troposphere exchange processes) as noted by Rao et al. (1997), Vukovich, (1997), Hogrefe et al. (2000), Porter et al. (2015), Astitha et al. (2017), Xing et al. (2016), and Mathur et al. (2017))."

Mathur, R.; Xing, J.; Gilliam, R.; Sarwar, G.; Hogrefe, C.; Pleim, J.; Pouliot, G.; Roselle, S.; Spero, T.L.; Wong, D.C.; Young, J. Extending the Community Multiscale Air Quality (CMAQ) modeling system to hemispheric scales: overview of process considerations and initial applications. *Atmos. Chem. Phys*. 2017, 17, 12449-12474, https://doi.org/10.5194/acp-17-12449-2017.

Xing, J., R. Mathur, J. Pleim, C. Hogrefe, J. Wang, C.-M. Gan, G. Sarwar, D. Wong, and S. McKeen, Representing the effects of stratosphere-troposphere exchange on 3D O_3 distributions in chemistry transport models using a potential vorticity based parameterization, Atmos. Chem. Phys., 16, 10865-10877, doi:10.5194/acp-16-10865-2016, 2016

Page 7, line 13: See my comment above: I couldn't fully understand how you constructed the "perfect model data". As far as I understood it, they would in any case only be perfect for this model setup and model grid. Could you comment on this?

We have revised the discussion in Section 3.2 to clarify that this analysis combining the observed baseline component with 21 CMAQ synoptic components is meant to quantify the amount of model error present in the current simulations that could conceivably be reduced through improving the representation of synoptic-scale processes and/or increased horizontal resolution. As part of this revision, we no longer refer to this combination of the measured baseline and modeled synoptic component as "perfect model". Moreover, we have added text at the end of the introduction to clarify that when we refer to the errors that can be expected even from a "perfect" model with "perfect" inputs throughout the manuscript, we consider these errors to be those arising from atmospheric stochasticity which we estimate in Section 3.1 using historic observations. We also should have noted in our original manuscript that four-dimensional data assimilation (FDDA) was utilized following the methodology suggested by Gilliam et al. (see *Atmos. Environ*., Vol.53, pp 186-201, 2012) and modified for fully-coupled meteorology-chemistry model applications as described in Hogrefe et al. (see *Atmos. Environ.*, Vol. 115, pp 683-694, 2015). As the reviewer pointed out, the SY component in the model output contains some meteorologically related variations on time scales of few days. The following revisions and additions have been made to address the reviewer's comment:

Revised and expanded the end of Section 1 (page 2, lines 29 – 32 in the original manuscript) as follows: "Also, no assessments have been made to date on the errors that are to be expected even from "perfect" regional-scale air quality modeling systems. To estimate such irreducible model errors due to atmospheric stochasticity (which we consider to be the errors that are expected even from a "perfect" model with "perfect" inputs), we analyzed the observed daily maximum 8-hr (DM8HR) ozone time series data at monitoring locations across the contiguous United States (CONUS) during the 1981-2014 time period and present the results of this analysis in Section 3.1. In Section 3.2, we illustrate how this information could be used in guiding model development specifically aimed at addressing reducible errors in the synoptic component by contrasting the results from Section 3.1 with analysis using the synoptic component from a 21-year simulation performed with the fully coupled WRF-CMAQ simulations covering the 1990-2010 period."

Expanded the model description in Section 2 (added after page 3 line 7 in the original manuscript) as follows: "To ensure better characterization of the prevailing meteorology (i.e., synoptic forcing) in the retrospective 21-year WRF-CMAQ simulations, four-dimensional data assimilation (FDDA) was utilized following the methodology suggested by Gilliam et al. (2012) and modified for fully-coupled meteorology-chemistry model applications as described in Hogrefe et al. (2015). The model set-up and performance evaluation of these historical multiyear WRF-CMAQ simulations have been published by Xing et al. (2015), Gan et al. (2015), and Astitha et al. (2017)."

Modified the discussion in Section 3.2 (page 5, line 23 – page 6, line 9 in the original manuscript) as follows: “In this section, we analyze long-term records of model simulations in an attempt to quantify the error associated with the modeled SY component that results both from not explicitly representing stochastic variations in atmospheric dynamics and emission variability in the current generation regional air quality models and from other reducible sources of model error. … To isolate the impact of model imperfections on only the SY time scale on errors across the ozone distribution, we assume that the model perfectly reproduces the ‘true’ BL depicted by the observed 2010 BL. We then use this ‘perfect’ modeled BL and reconstruct ‘pseudo-simulated’ ozone time series, similar to what was done in Fig. 3, except for using the SY component embedded in the 21 years of coupled WRF-CMAQ simulations. The rationale for this analysis is to quantify the amount of model error present in the current simulations that could conceivably be reduced through improving the representation of synoptic and mesoscale processes and/or increased horizontal resolution with appropriate data assimilation techniques.”

Caption of Figure 2: Explain the meaning of the number 420130801. Explain that the “observed” line in Fig 2a is for 2010.

The number represents the site # in EPA’s AQS database; it has no specific meaning other than being an identifier for the location of the monitoring site.

The figure caption (Figure 2 in the original manuscript, Figure 3 in the revised manuscript) has been updated as follows: “Figure 3a: Comparison between the observed cumulative distribution function (CDF) for 2010 shown in red with 30+ pseudo-observations CDFs generated from historical DM8HR ozone time series shown in gray at a suburban site at Altoona in PA (AQS station identifier 420130801). The blue line represents the average of the 30+ gray lines; Figure 3b: Display of various statistical metrics (standard deviation (std), root mean square error (RMSE), bias) derived by comparing the actual observed and pseudo ozone values in Fig. 3a; Figure 3c: Normalized statistical metrics of normalized mean error (NME), normalized mean bias (NMB), coefficient of variation (CV). Notice the large variability occurring at the lower and upper percentiles”

Caption of Figure 3: Give the equations you refer to somewhere in this paper (e.g. in an appendix).

The paper by Emery et al. (2016) and many other papers included in the references list recommended the statistical metrics such as RMSE, NME, Bias, NMB, CV, etc. whose definitions are well known.

Updated the second paragraph of Section 3.1 (page 5 line 9 of the original manuscript) in which Figure 3 (Figure 2 in the original manuscript) is discussed as follows: “see Emery et al. (2016) for the description of the statistical metrics considered here”

Figure 5: Use same colors for observations and model in both graphs.

Done.

Caption of Figure 6: “5)” should be “c)”. “Light blue” in Fig 6d) appears to be grey. Figure 4 and Figure 7: Give units (ppb).

Done.

Authors' Response (in red) to Dr. William Stockwell's Comments

Rao et al. examine how the stochastic variability of the atmosphere affects the accuracy of regional air quality model predictions. Stochastic variability would be expected to introduce error in predictions even if the model is "perfect". This paper provides an analysis of the expected error. The question of the limits of "predictability" of models is well known in meteorology but it has not been explored very extensively for air quality models. Therefore, this paper provides a valuable contribution to the literature. The paper provides an excellent basis for future research and improvements to air quality models as well.

We thank Dr. Stockwell for his positive feedback on our paper.

Atmospheric stochastic variability extends to scales that are well below current Eulerian model resolution. Eulerian models calculate gas-phase chemical transformations across the modeling domain within grid-boxes and instantaneous uniform mixing of chemical species is assumed for each grid-box. However, the stochastic variability of wind fields suggests that chemical concentrations should be represented by mean and varying components. This difference between reality and model representation may be most important for rapid, diffusion-limited reactions that affect ozone and particulate formation (Stockwell, J. of Meteor. and Atmos. Phys., 57, 159-172, 1995).

Agreed. We hope that the next generation of operational Eulerian models would be able to handle the physical and chemical processes as suggested by Dr. Stockwell. Also, it is important to resolve emissions inventories to the time and space scales of the model.

On the Limit to the Accuracy of Regional-Scale Air Quality Models

S. Trivikrama Rao[a,b], Huiying ~~Luo~~[a]Luo[b], Marina ~~Astitha~~[a]Astitha[b], Christian Hogrefe[c], Valerie Garcia[c], Rohit Mathur[c]

[a]Department of Marine, Earth, and Atmospheric Sciences, North Carolina State University, Raleigh, NC
[b]Department of Civil and Environmental Engineering, University of Connecticut, Storrs, CT
[c]~~Computational Exposure Division~~Center for Environmental Measurement & Modeling, U.S. Environmental Protection Agency, Research Triangle Park, NC

Correspondence to: S. Trivikrama Rao (strao@ncsu.edu)

Abstract. Regional-scale air pollution models are routinely being used world-wide for research, forecasting air quality, and regulatory purposes. It is well known that there are both reducible and irreducible uncertainties in the meteorology-atmospheric chemistry modeling systems. Inherent or irreducible uncertainties stem from our inability to properly characterize stochastic variations in atmospheric dynamics and from the incommensurability associated with comparisons of the volume-averaged model estimates with point measurements. Because stochastic variations in atmospheric dynamics and emissions forcing influencing the air pollutant concentrations are difficult to explicitly simulate, one can expect to find a percentile value from the distribution of measured concentrations to have much greater variability than that of the model. This paper presents an observation-based methodology to determine the expected errors from regional air quality models even when the model design, physics, chemistry, and numerical analysis techniques as well as its input data were "perfect". To this end, the short-term synoptic-scale fluctuations embedded in the daily maximum 8-hr ozone time series are separated from the longer-term ~~forcings~~forcing using a simple recursive moving average filter. The inherent variability attributable to the stochastic nature of the atmosphere is determined based on 30+ years of historical ozone time series data measured at various monitoring sites in the contiguous United States. The results reveal that the expected root mean square error at the median and 95th percentile is about 2 ppb and 5 ppb, respectively, even for "perfect" air quality models driven with "perfect" input data. Quantitative estimation of the limit to the model's accuracy will help in objectively assessing the current state-of-the-science in regional air pollution models, measuring progress in their evolution, and providing meaningful and firm targets for improvements in their accuracy relative to ambient measurements.

1 Introduction

Confidence in model estimates of pollutant distributions is established through direct comparisons of modeled concentrations with corresponding observations made at discrete locations for retrospective cases. It is well known that there are both reducible and irreducible uncertainties in the meteorology-atmospheric chemistry modeling systems. Pinder et al. (2008) discussed the

reducible (i.e., structural and parametric) uncertainties that are attributable to the errors in model input data (e.g., meteorology, emissions, initial and boundary conditions) as well as our incomplete or inadequate understanding of the relevant atmospheric processes (e.g. chemical transformation, planetary boundary layer evolution, transport and dispersion, modeling domain, grid resolution, deposition, rain, clouds). Inherent or irreducible uncertainties stem from our inability to properly characterize the stochastic variations in atmospheric dynamics (~~Gilliam~~Rao et al., ~~2015~~1985; Rao et al., , 2011), from the incommensurability associated with comparing the volume-averaged model estimates with point measurements (e.g., McNair et al., 1996; Swall and Foley, 2009), and our inability to precisely quantify the space and time variations in atmospheric emissions and other atmospheric variables and processes. Also, without completely knowing the 3-dimensional initial physical and chemical state of the atmosphere, its future state cannot be simulated accurately (Lamb, 1984; Lamb and Hati, 1987; Lewellen and Sykes, 1989; Pielke, 1998; Gilliam et al., 2015). Given the presence of the irreducible uncertainties, precise replication of observed concentrations or their changes by the models cannot be expected (Dennis et al., 2010~~).~~; Rao et al., 2011; Porter et al., 2015).

Whereas an air quality model's prediction represents some time/space-averaged concentrations, an observation at any given time at a monitoring location reflects an individual event or specific realization out of a population that will almost always differ from the model estimate even if the model and its input data were perfect (Rao et al., 1985). Consequently, comparisons of modeled and observed concentrations paired in space and time indicate biases and errors in simulating absolute levels of pollutant concentrations at individual monitoring sites (Porter et al., 2015). The scientific discussion on modeling uncertainty reduction goes back more than three decades with the current practice including data assimilation, ensemble modeling, and model performance evaluation (e.g., Fox, 1981, 1984; Lamb, 1984; Pielke, 1998; Lewellen and Sykes, 1989; Lee et al., 1997; Carmichael et al., 2008; Hogrefe et al., 2001a, 2001b; Grell and Baklanov, 2011; Gilliam et al., 2006; Baklanov et al., 2014; Bocquet et al., 2015~~).~~; Solazzo and Galmarini, 2015). While ever-improving process knowledge and increasing computational power will continue to help reduce the structural and parametric uncertainties in air quality models, the inherent uncertainty cannot be eliminated because our inability to properly characterize the stochastic nature of the atmosphere will always result in some mismatch between the model results and measurements; this could lead to speculation on the inferred accuracy of the future states simulated by the regional-scale air quality models (Dennis et al., 2011; Rao et al., 2011; Porter et al., 2015; Astitha et al., 2017; Luo et al., 2019).

In most applications of regional-scale air quality models, statistical metrics such as bias, root mean square error (RMSE), correlation, and index of agreement are being used to judge the quality of model predictions and determine if the model is suitable for forecasting or regulatory purposes (e.g., Fox, 1981, 1984; Solazzo et al., 2011; Appel et al., 2012; Simon et al., 2012; Foley et al., 2014; Ryan et al., 2016; ~~Emory~~Emery et al. 2016; Zhang, 2016; U.S. EPA, 2018). While significant improvements in the formulation, physical and chemical parameterizations, and numerical techniques have been implemented in atmospheric models over the past three-decades, it is not clear if the improvement claimed in the model's performance relative to the routine network measurements is statistically significant based on these metrics (Hogrefe et al., 2008). Also, no

assessments have been made to date on the errors that are to be expected ~~in~~even from "perfect" regional-scale air quality ~~models.~~modeling systems. To ~~this end,~~estimate such irreducible model errors due to atmospheric stochasticity (which we consider to be the errors that are expected even from a "perfect" model with "perfect" inputs), we analyzed the observed daily maximum 8-hr (DM8HR) ozone time series data at monitoring locations across the contiguous United States (CONUS) during the 1981-2014 time period ~~along~~and present the results of this analysis in Section 3.1. In Section 3.2, we illustrate how this information could be used in guiding model development specifically aimed at addressing reducible errors in the synoptic component by contrasting the results from Section 3.1 with ~~the~~ analysis using the synoptic component from a 21-year simulation performed with the fully coupled WRF-CMAQ simulations covering the 1990-2010 period ~~as detailed below.~~

2 Data and Methods

Ground-level DM8HR ozone data covering the CONUS during May to September in each year were obtained from the U.S. Environmental Protection Agency's (EPA) Air Quality System (AQS) (see https://www.epa.gov/aqs). A valid ozone season consists of at least 80% data coverage during May to September at each station. A total 185 monitoring stations with at least 30 valid years (to provide enough variety of synoptic conditions, denoted hereafter as 30+ in this paper) from the year 1981 to 2014 are analyzed. Also, fully coupled WRF-CMAQ model simulations over the CONUS for the 1990-2010 period were utilized in this study to demonstrate a new perspective on model performance evaluation. To ensure better characterization of the prevailing meteorology (i.e., synoptic forcing) in the retrospective 21-year WRF-CMAQ simulations, four-dimensional data assimilation (FDDA) was utilized following the methodology suggested by Gilliam et al. (2012) and modified for fully-coupled meteorology-chemistry model applications as described in Hogrefe et al. (2015). The model set-up and performance evaluation of these historical multiyear WRF-CMAQ simulations have been published by Xing et al. (2015), Gan et al. (2015), and Astitha et al. (2017). Time-varying chemical lateral boundary conditions are nested from the 108 km hemispheric WRF-CMAQ simulation from 1990 to 2010 (Xing et al., 2015). ~~Evaluation of the 21-year long WRF-CMAQ simulation using 36-km grid can be found in Gan et al. (2015).~~

It has been shown that time series of the daily maximum 8-hr ozone concentrations contain fluctuations operating on different time scales~~, reflecting the short-term~~ (e.g., intra-day forcing induced by the fast-changing emissions and atmospheric boundary layer evolution; diurnal forcing induced by the day and night differences; synoptic forcing induced by the passage of weather systems across the country, sub-seasonal forcing due to Madden-Julian Oscillation (MJO), and long-term forcing induced by emissions, El-Nino-Southern Oscillation (ENSO), climate change, and other slow-varying processes such as seasonal and sub-seasonal variations in the atmospheric deposition and stratosphere-troposphere exchange processes ~~(~~) as noted by Rao et al.~~,~~ ~~1996,~~ (1997~~;~~), Vukovich, (1997~~;~~), Hogrefe et al.~~,~~ (2000~~;~~), Porter et al.~~,~~ (2015~~;~~), Astitha et al.~~,~~ (2017~~).~~), Xing et al. (2016), and Mathur et al. (2017)). Variations in ~~ambient~~8-hour ozone can be thought of comprising of the baseline of pollution that is created by various emitting sources and modulated by the prevailing synoptic weather conditions (Rao et al., 2011). Thus,

the level of the baseline (BL) concentration and the strength of the synoptic component (SY) should be viewed as the necessary and sufficient conditions for how high ozone levels can reach on a given day (Astitha et al., 2017). Scale separation can be achieved by applying filtering methods such as the Empirical Mode Decomposition (Huang et al., 1998), Elliptic filter (Poularika, 1998), ~~Kolomogorov~~Kolmogorov-Zurbenko (KZ) filter (Rao and Zurbenko, 1994), Adaptive Filter Technique (Zurbenko, et al., 1996), and Wavelet (Lau and Weng, 1995). Because Improved Complete Ensemble Empirical Mode Decomposition with Adaptive Noise (Improved CEEMDAN, a version of the Empirical Mode Decomposition) and KZ filter yielded similar results for the DM8HR time series data as shown in Figs. 1-2 discussed in the next section, only the results from the KZ filter are presented in the subsequent analysis for quantifying the impact of the stochastic nature of the atmosphere on observed and simulated ozone concentrations. Furthermore, the KZ filtering is a simple method and works well even in the presence of missing data (Hogrefe et al., 2003~~),~~). In this study, we used the ~~KZ(~~$KZ_{5,}$5~~,5) in this study~~ with a window size of 5 days and 5 iterations on raw ozone time series [O_3 (t)] in the same manner as in Porter et al. (2015), Rao et al. (2011), and Luo et al. (2019). The size of the window and the number of iterations determine the desired scale separation. The ~~KZ(~~$KZ_{5,}$5~~,5)~~ filtering process helps separate the synoptic-scale weather-induced variations embedded in the May-September DM8HR time series data (short-term component, noted as SY) from the long-term baseline component (noted as BL). ~~It should be noted that the application of the Empirical Mode Decomposition (EMD) and KZ (5,5) to the DM8HR time series data yielded similar results.~~

$$BL(t) = \sout{KZ(5,5)}\ KZ_{5,5}\left(O_3(t)\right) \quad (1)$$

$$SY(t) = O_3(t) - \sout{KZ(5,5)}\ KZ_{5,5}\left(O_3(t)\right) \quad (2)$$

$$O_3(t) = SY(t) + BL(t) \quad (3)$$

~~where O3(t) is the original time series of the observed DM8HR ozone concentration, BL(t) is the baseline component and SY(t) is the synoptic component at any given time.~~ Because we are working with the daily maximum 8-hr ozone data, the Nyquist interval is 2-days, indicating that the dynamical features having time scales less than 2 days (e.g., intra-day forcing from fast changing emissions and chemical transformations, boundary layer evolution, diurnal forcing due to night vs. day differences) are not resolvable in this analysis (see Fig. 2 in Dennis et al., 2010). The 50% cut-off frequency for the ~~KZ(~~$KZ_{5,}$5~~,5)~~ is ~24 days, and, hence, time scales less than those associated with synoptic-scale weather fluctuations are embedded in the short-term or SY forcing. The KZ filtering is applied to both DM8HR observations and modeled DM8HR time series. Once the baseline is separated from the original DM8HR time series from all monitoring stations, then the synoptic forcing in the historical ozone time series data is used to estimate the variability in ozone concentrations that can be expected because of the chaotic/stochastic nature of the atmosphere by taking into account the relationship between the strength of synoptic forcing and mean of baseline ozone at each location over CONUS. This methodology was applied to both measured and modeled ozone concentrations (see details in Luo et al., 2019). Whereas the objective of Luo et al. (2019) was on transforming the deterministic modeling results into a probabilistic framework for assessing the efficacy of different emission control strategies in achieving compliance with the ozone standard, this paper is aimed at quantifying the model performance errors to be

expected at each monitoring site over CONUS even from "perfect" regional ozone models driven with "perfect" input data from the ever-present stochastic nature of the atmosphere.

3 Results and Discussion

3.1 Analysis of ambient ozone data

Using both Improved CEEMDAN and KZ filtering, we separated the synoptic forcing (time scale < 24 days) and baseline (time scale > 1 month) forcing embedded in the time series of observed and modeled daily maximum 8-hour ozone concentrations. To illustrate, the results from the application of Improved CEEMDAN to the daily maximum 8-hr ozone time series data measured at Altoona, PA are presented in Fig. 1. The top left panel displays the raw ozone time series while the top of the right panel shows its power spectrum. The 7 intrinsic mode functions (IMFs) and the residual on the left side, and their corresponding power spectra on the right reveal that most of the synoptic-scale features in ozone data are imbedded in IMFs 1 and 2. The baseline ozone is extracted by removing the first two IMFs from the raw ozone time series. To illustrate the concept of the ozone baseline, DM8HR time series measured in 2010 at Altoona, PA is presented in Fig. ~~1a~~2a together with the embedded baseline concentration as extracted by the ~~KZ(~~$KZ_5$5~~,5) filter~~ and Improved CEEMDAN. It is evident that high ozone levels are always associated with the elevated baseline. The difference between the raw ozone time series and baseline, denoted as the short-term or synoptic forcing (SY), is displayed in Fig. ~~1b along with time series of white noise. By superimposing AR(1) process on the ozone baseline, Rao et al. (1996) demonstrated that the number of observed ozone exceedances above a given threshold at a monitoring site can be reproduced. A comparison between the SY component and white noise process, presented in Fig. 1b, reveals that the SY component having finite variance and zero mean resembles near-stochastic process. Hence, the baseline concentration is to be viewed as the deterministic part and SY is considered the stochastic component in the ambient ozone time series~~2b. The power spectra, displayed in Figs. 2c and d, reveal both methods yielded good scale separation. Due to the good agreement between both scale separation techniques, only the results from the KZ filter are presented for the remainder of the manuscript.

Once the scale separation is achieved with the ~~KZ(~~$KZ_5$5~~,5),~~ we superimposed the SY forcing imbedded in 30+ years of historical DM8HR ozone time series measured at a given location on the baseline component of the ozone time series at that location to generate 30+ reconstructed or pseudo ozone distributions. Illustrative results using eq. (3) at a suburban location in Altoona, PA are presented for 2010 base year in Fig. ~~2a~~3a; it should be noted that the linear relationship between the strength of SY (defined as the standard deviation of the data in the synoptic component) and the magnitude of the BL (defined as the mean of the data in the baseline component) has been taken into account in generating 30+ years of adjusted SY forcing as illustrated in Luo et al. (2019). As expected, there is excellent agreement between the average of 30+ values (solid blue line) and observed ozone in 2010 at each percentile of the concentration distribution function (red line). Also, the original cumulative distribution function (CDF) in 2010 (red line) is constrained within the 30+ CDFs of pseudo ~~observations~~ distributions (Fig.

~~2a~~3a); note, it is equally likely for any of ~~the~~these 30+ CDFs to occur ~~due to~~because of the stochastic nature of the atmosphere even though the individual event in 2010 yielded the CDF shown in red. As mentioned before, ozone mixing ratio at any given probability point on the red line in Fig. ~~2a~~3a reflects ~~a specific~~an individual event while ozone values at the same probability in different CDFs (~~light blue~~gray lines) reflect the population stemming from the ~~chaotic~~stochastic nature of the atmosphere. In other words, there are 30+ dynamically consistent ozone time series attributable to the 2010 baseline (given 2010 emissions ~~loading)~~ for examining the inherent variability~~.~~ due to atmospheric stochasticity. It is evident in Fig. ~~2a~~3a that there is larger variability at the lower and upper percentiles than that in inter-quartile range, revealing that the tails of the concentration distribution function are subject to large inherent uncertainty. Using these 30+ pseudo-observation ozone mixing ratios and the actual observed ozone values at each percentile, statistical metrics such as Bias, RMSE, coefficient of variation (CV=standard deviation/mean), normalized mean error (NME) and normalized mean bias (NMB) are presented in Fig. ~~2b and c.~~3b and c (see Emery et al. (2016) for the description of the statistical metrics considered here). As expected, the lower and upper tails of the distribution are prone to large errors. These results demonstrate the presence of ~~larger~~substantial natural variability at the upper 95th percentile, which is of primary interest in regulatory analyses. The extreme values are better described in statistical terms rather than in deterministic sense (Hogrefe and Rao, 2001).

Ozone time series at 185 monitoring stations covering CONUS, having at least 80% data completeness, are analyzed in the above manner and the results are displayed as box plots in Fig. ~~3~~4. Note the presence of large variability in the CV, NME, and NMB, and Bias at the lower and upper percentiles (Fig. ~~3~~4). The RMSE expected for the ozone mixing ratios in the interquartile range is ~1.5 ppb, but it is >5 ppb for the upper 95th percentile (Fig. ~~3b~~4b). The spatial distribution of RMSE at the 50th and 95th percentiles is displayed in Figures ~~4a~~5a and ~~4b~~5b, respectively. The RMSE at the upper 95th percentile is very high at some monitoring sites in California and Michigan (Fig. ~~4b~~5b). Monitoring stations ~~at high elevations, residing well above the nocturnal boundary layer, tend to exhibit lower variability than those~~ situated in the urban areas, near large water bodies, and in regions of complex terrain ~~due to the dominance of~~ influenced predominantly by local conditions tend to exhibit higher RMSE. The elevation of the monitoring sites is displayed in Fig. 5c.

3.2 Analysis of modeled ozone concentrations

The analysis in the previous section quantified the inherent stochastic variability ~~represented by~~that is present in the SY component using long-term records of ozone observations. In this section, we analyze long-term records of model simulations in an attempt to quantify the error associated with the modeled SY component that results both from not explicitly representing stochastic variations in atmospheric dynamics and emission variability in the current generation regional air quality models~~.~~ and from other reducible sources of model error. The model simulations were performed with the fully coupled WRF-CMAQ system with a 36-km horizontal grid cell size and covered the 21-year period from 1990 to 2010 (Gan et al., 2015). To provide an illustration of the differences between observed and modeled time series over this period, Figure ~~5a~~6a displays a scatter plot of the strength of the SY component ~~vs.~~(standard deviation of data in the SY component) vs. the mean of the baseline

(BL) component for both observations and model simulations at the Altoona, PA site. While both observations and WRF-CMAQ simulations show a strong correlation between these two variables, it is evident that at this monitoring location the standard deviation (i.e., strength) of the SY component is substantially lower for the WRF-CMAQ simulations for a given mean of the BL component (i.e., for any given year). The year-to-year variation in the observed and modeled mean of BL and strength of SY forcing, displayed in Fig. ~~5b~~6b, reveals that the model overestimated BL and underestimated the strength of SY forcing. The 36-km grid may be better ~~reproducing~~representing the large-scale synoptic forcing associated with the translation of weather systems than the meso-scale weather and urban influences (both dynamics and emission driven) that are embedded in the observed SY component. Meteorological modeling with higher horizontal grid resolution might be able to capture the land-sea breeze, lake-sea breeze, and terrain influences that observations are seeing at certain monitoring locations.

~~An understanding~~To isolate the impact of ~~the expected error even when the model's physics, chemistry, numerical solver, and the input data are "perfect" would help~~ model ~~developers in making decisions~~imperfections on ~~model improvements. To this end~~only the SY time scale on errors across the ozone distribution, we assume that the model perfectly reproduces the 'true' BL depicted by the observed 2010 BL. We then use this 'perfect' modeled BL and reconstruct 'pseudo-simulated' ozone time series, similar to what was done in Fig. ~~2~~3, except for using the SY component ~~from~~embedded in the 21 years of coupled WRF-CMAQ simulations. The rationale for this analysis is to quantify the amount of model error present in the current simulations that could conceivably be reduced through improving the representation of synoptic and mesoscale processes and/or increased horizontal resolution with appropriate data assimilation techniques. Fig. ~~6a shows~~7a displays the CDF of actual observed ozone (red line) overlaid on 21 pseudo-simulated ozone CDFs (~~blue~~gray lines, with averages of all 21 pseudo-simulated ozone percentiles shown in blue) at the Altoona, PA site while Figs. ~~6b~~7b and ~~6c~~7c display absolute and normalized performance metrics. Figure ~~6a~~7a confirms that the coupled WRF-CMAQ SY components have less intra-annual ~~(sub-seasonal)~~ variability than observed SY components, causing ~~an~~ overestimation at the low end and ~~an~~ underestimation at the high end of the observed CDF for all 21 years of reconstruction; these results imply that the model's results at the upper and lower percentiles will always tend to be unreliable or prone to large errors even when the baseline concentration is predicted perfectly. The U-shape of the absolute and relative error curves in Figures ~~6b~~7b and c is similar to the corresponding curves in Figure ~~2~~3, but the larger magnitude at the high and low end of the distribution indicates that the effects of the underestimated intra-annual ~~(sub-seasonal)~~ SY variability (note that the distribution of modeled values in Fig. ~~6a~~7a is much flatter (i.e., having higher Kurtosis) than that of the observations) outweigh those errors attributable to the stochastic variability presented in Figure ~~2~~3. The shape of the absolute and normalized bias curves deviates from those shown for the pseudo-observations in Figures ~~2b~~3b-c and, thus, also reveals the effect of the underestimation of the ~~sub-seasonal~~intra-annual SY variability. Figures ~~6d~~7d-f present differences between the curves shown in Figures ~~6a~~7a-c and a version of Figure ~~2a~~3a-c computed from the 1990-2010 ~~rather than~~ data instead of 30+ years of historical ozone observations. Panels e and f show that at the 50th percentile, the differences in the error curves are close to zero due to the fact that both the pseudo-simulations and pseudo-observations used the same observed BL component. At the upper percentiles, the differences reach 3 – 5 ppb, providing an estimate of the

reducible error in simulating the extreme values at this location because of the differences in the observed and WRF-CMAQ SY components at this location; high-resolution meteorological modeling may help address these reducible errors.

Figs. ~~7a~~8a and ~~7b show~~b display the RMSE at the median and 95th percentile for the 'pseudo-simulated' ozone values at each monitoring site. For the 50th percentile, the RMSE values range from 0.2 ppb to 3.2 ppb over CONUS with a median value of 1 ppb while at the 95th percentile, the RMSE values range from 1 ppb to ~~14.9~~15 ppb with a median value of ~~3.8~~4 ppb across all sites over CONUS. The values are highest along the California coast and near Great Lakes, possibly due to ~~errors in boundary conditions and~~ inadequacies in simulating the land-sea breeze and land-lake breeze regimes, respectively, with modeling at 36 km ~~grids.~~ grid cells. Air quality modeling uncertainty even for the retrospective modeling cases, outside of the chemistry formulation and boundary conditions, is attributed primarily to meteorology and emissions inputs. Vautard et al. (2012) concluded that major challenges still remain in the simulation of prevailing meteorology (e.g., errors in wind speed, PBL, night-time meteorology, clouds) in retrospective air quality modeling. Based on the retrospective ozone episodic modeling with the WRF-CMAQ model using various sets of equally likely initial conditions for meteorology along with FDDA, Gilliam et al. (2015) confirmed the presence of sizable spread in WRF solutions, including common weather variables of temperature, wind, boundary layer depth, clouds, and radiation, thereby causing a relatively large range of ozone concentrations. Also, pollutant transport is altered by hundreds of kilometers over several days. Ozone concentrations of the ensemble varied as much as 10–20 ppb (or 20–30%) in areas that typically have higher pollution levels. As model improvements are made, one can quantitatively assess how close the predictions of the improved model are ~~to the expected or target RMSE at each monitoring site~~ for each percentile for the given base year simulation ~~(see Fig.4a and b for~~to the expected errors from a "perfect" ~~models~~model with "perfect" input ~~at~~, i.e. the ~~median~~target RMSE shown in Fig.5a and ~~95th percentile).~~b.

4 Conclusions

~~Weather is a stochastic process that impacts the prediction of air pollutants, and regardless~~Regardless of how accurate the regional air quality model is, ~~this~~the stochastic ~~component~~variations in the atmosphere cannot be consistently reproduced by the deterministic numerical models. In this study, we demonstrate how to quantify this irreproducible stochastic component by isolating the synoptic forcing imbedded in 30+ years of historical observations and assess the performance of the 36 km fully coupled WRF-CMAQ model in simulating 21 years of ozone concentrations over ~~CONUS~~the contiguous U.S. Observation-based analysis reveals that on average, the irreducible error attributable to the stochastic nature of the atmosphere ranges from ~2 ppb at the 50th percentile to ~ 5 ppb at the 95th percentile. To improve regional-scale ozone air quality models, attention should be paid to accurately simulate the baseline concentration by focusing on the quality of the emission inventory and the model's treatment for the boundary conditions and slow-changing (operating on sub-seasonal, seasonal, and longer-term time scales) atmospheric processes. Also, errors in reproducing the ~~sub-seasonal variability~~synoptic forcing can possibly be reduced with high-resolution meteorological modeling using appropriate data assimilation techniques. Nonetheless, these

results demonstrate the presence of large variability in the upper tail of the DM8HR O_3 concentration cumulative distribution even with "perfect" models using "perfect" input data. Having this quantitative estimation of practical limits for model's accuracy helps in objectively assessing the current state of regional-scale air quality models, measuring progress in their evolution, and providing meaningful and firm targets for improvements in their accuracy relative to measurements from routine networks.

Code availability: Source code for version 5.0.2 of the Community Multiscale Air Quality (CMAQ) modeling system can be downloaded from https://github.com/USEPA/CMAQ/tree/5.0.2. For further information, please visit the U.S. Environmental Protection Agency website for the CMAQ system: https://www.epa.gov/cmaq.

Data availability: All ozone observations used in this article are available from https://aqs.epa.gov/aqsweb/airdata/download_files.html (AQS). Paired ozone observation and CMAQ model data used in the analysis will be made available at https://edg.epa.gov/metadata/catalog/main/home.page. Raw CMAQ model outputs are available on request from the U.S EPA authors.

Competing interests: The authors declare that they have no conflict of interest.

Author Contribution: STR conceptualized the idea. STR, CH, VG, and RM designed the analysis approach. CH and RM post-processed previously conducted model simulations. HL performed data analyses and prepared the illustrations. STR prepared the manuscript with contributions from all co-authors.

Disclaimer: The views expressed in this paper are those of the authors and do not necessarily represent the view or policies of the U.S. Environmental Protection Agency.

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Figure 6. (a) Scatter plot of the standard deviation (i.e., strength) of the SY component vs. the mean of the baseline (BL) component for each of the 21 years from 1990 to 2010 at the Altoona, PA monitoring site. ~~Figure 5. (a) Scatter plot of the standard deviation (i.e., strength) of the SY component vs. the mean of the baseline (BL) component for each of the 21 years from 1990 to 2010 at the Altoona, PA monitoring site.~~ Observations are shown in ~~blue~~red while WRF-CMAQ results are shown in ~~red~~blue. (b) Inter-annual variability in the mean of the baseline component and standard deviation of the synoptic component in the WRF-CMAQ model and observations at the Altoona, PA site. Although year-to-year variation is captured, the model has overestimated the baseline forcing and underestimated the synoptic forcing.

Figure ~~6~~7. a) Comparison between the observed CDF ~~overlain~~overlaid on 21 'pseudo-simulated' or reconstructed ozone CDFs with SY generated from modeled DM8HR ozone time series at a suburban site ~~(420130801)~~ at Altoona in~~;~~ PA (AQS station identifier 420130801); b) Display of various statistical metrics derived by comparing the actual observed and pseudo-simulated ozone values in Fig. ~~6a; 5~~7a; c) Normalized statistical metrics; d).Difference between the pseudo-simulated CDFs shown in Figure ~~6a~~7a and the pseudo-observed CDFs as shown in Figure ~~2a~~7a but calculated from 21 years (1990-2010) of observations only. The ~~light blue~~gray lines represent the differences for a specific SY year while the ~~thick~~ blue line represents the differences between the means of the 21 reconstructions; e) Difference between the absolute performance metrics for pseudo-simulations shown in Figure ~~6b~~7b and those calculated for pseudo-observations as shown in Figure ~~2b~~7b but calculated for 21 years (1990-2010) only. f) As in panel e) but for normalized performance metrics.

Figure ~~7~~8. Errors ~~attributable to the different synoptic forcings~~ in ~~model results~~ the 21 'pseudo-simulated' or reconstructed ozone time series with SY generated from modeled DM8HR ozone time series using BL obtained from observations at (a) the median and (b) 95th percentile.

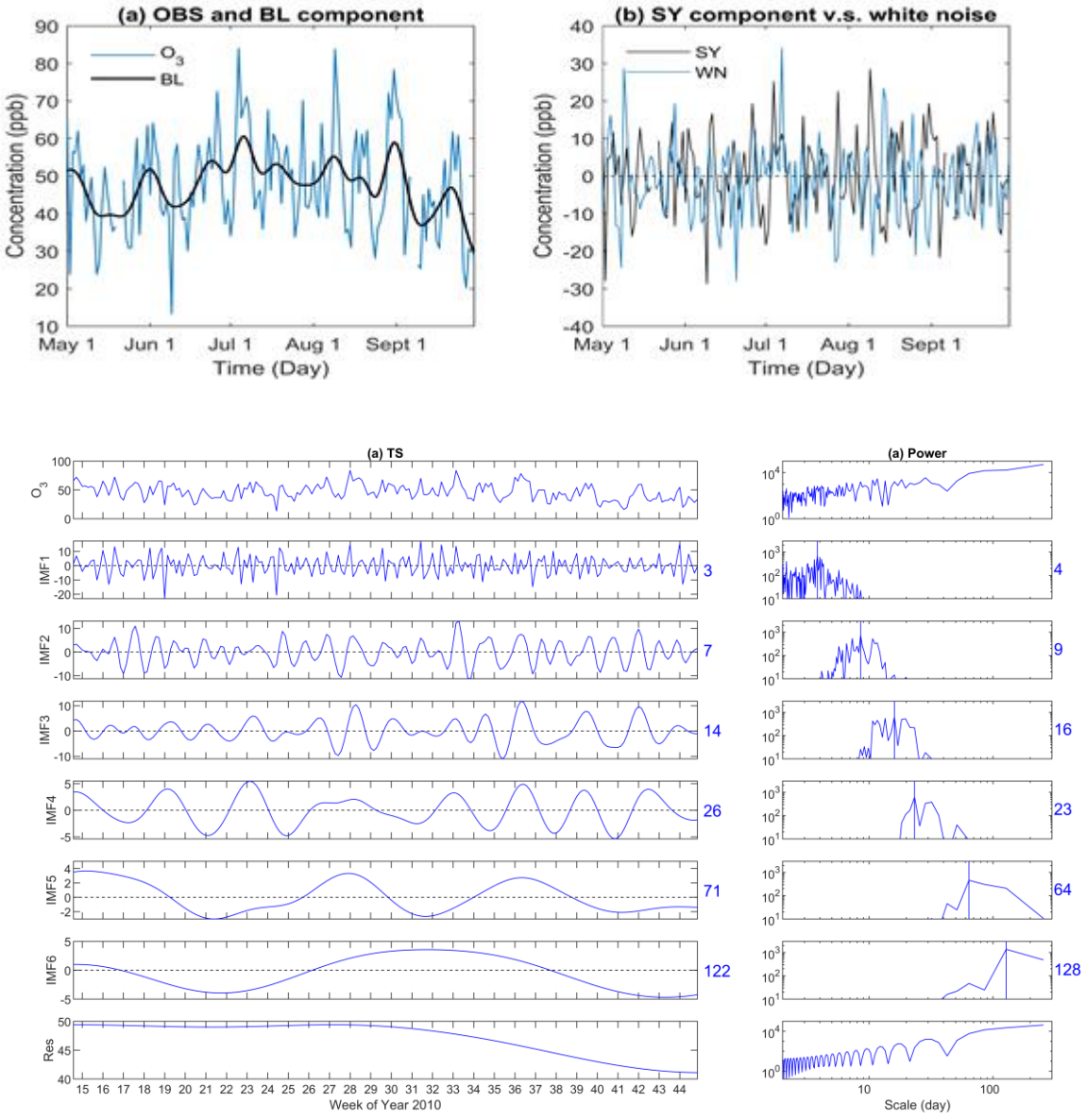


Figure ~~1a. Observed~~1. Results of the application of the Improved CEEMDAN technique (a modified version of EMD) technique, which is designed for analyzing non-stationary and non-linear time series data, to the daily maximum 8-hour ozone time series data at the Altoona, PA site. The numbers on the right side represent the time scale (in days) associated with each IMF. Note, the power spectrum of raw ozone time series (upper right panel) shows that the energy in the 1-10 days (synoptic) time scale is an order of magnitude less than that in the longer (baseline) time scale.

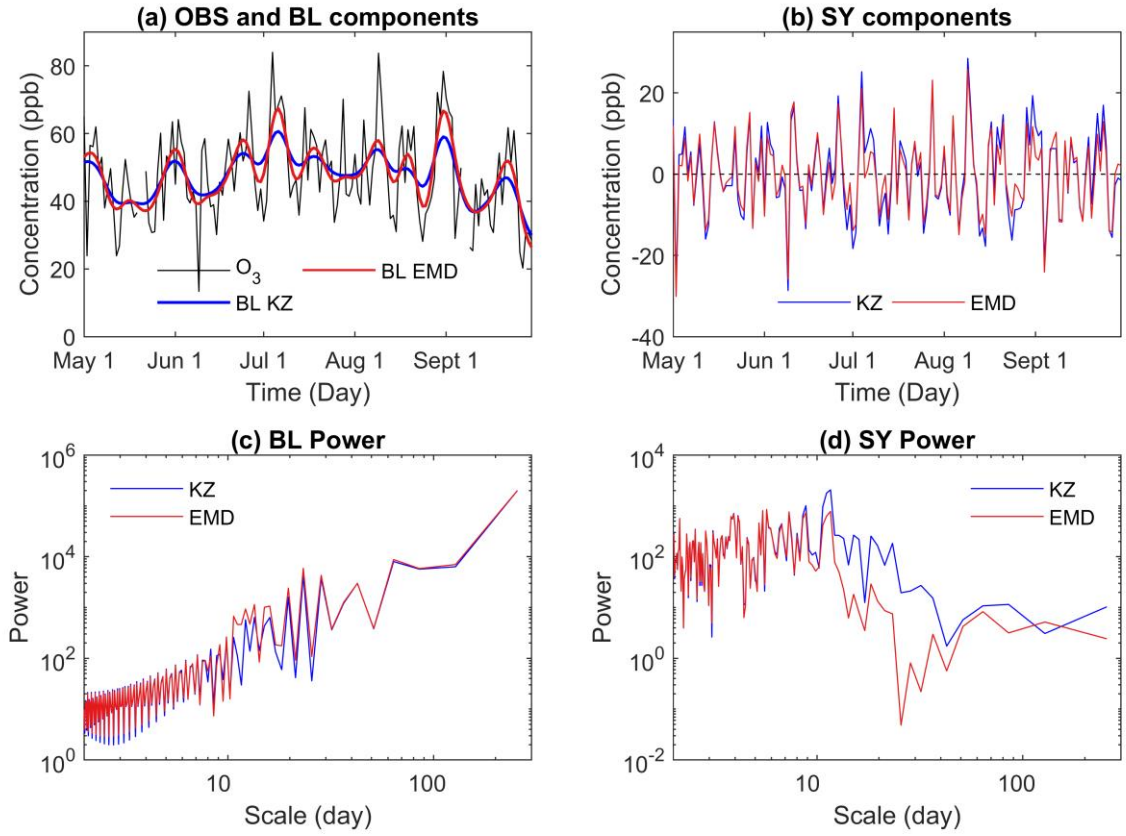
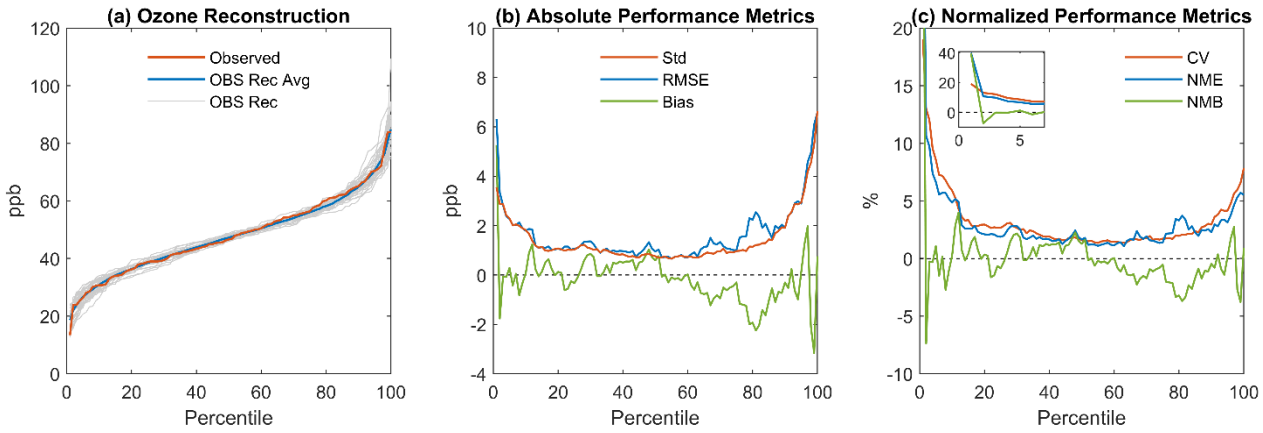


Figure 2a. Raw observed DM8HR ozone time series (~~blue line~~black) and the embedded baseline (~~black line~~red for EMD and blue for KZ) at Altoona, PA in 2010; Figure ~~1b~~2b. Time series of synoptic forcing (~~black line) and time series of Gaussian white noise (~~red for EMD and blue ~~line) having the same variance as SY forcing~~for KZ); Figure 2c and 2d are their corresponding power spectra. The bottom two panels compare the power spectra of the baseline forcing (left) and the synoptic forcing (right) derived from KZ filtering and EMD (sum of IMF 1 and IMF2). Notice that most of the energy in the baseline time series is in the longer time scale while most of the energy of the short-term component is in the high-frequency range. The similarity of results from both scale separation techniques demonstrates that the two scales of interest (i.e., baseline and synoptic forcing) have been extracted reasonably well by these two methods.


(a) Ozone Reconstruction
Observed
OBS Rec Avg
OBS Rec
ppb
Percentile
(b) Absolute Performance Metrics
Std
RMSE
Bias
ppb
Percentile
(c) Normalized Performance Metrics
CV
NME
NMB
%
Percentile

5

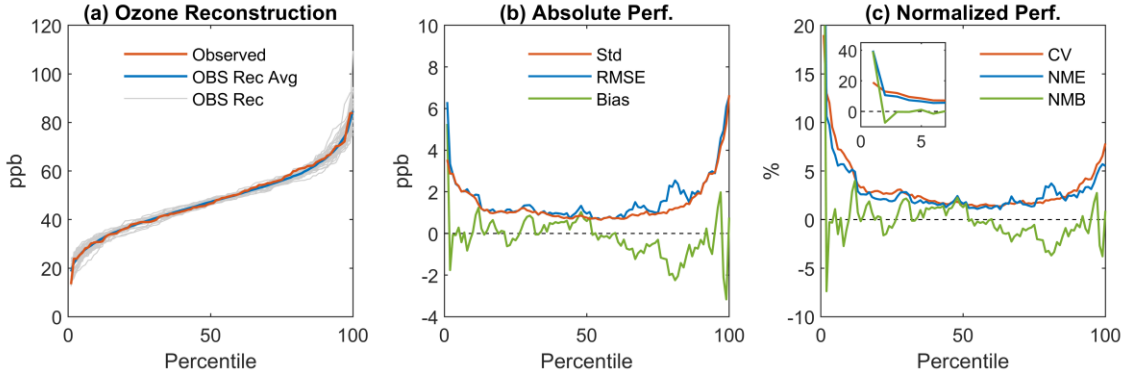
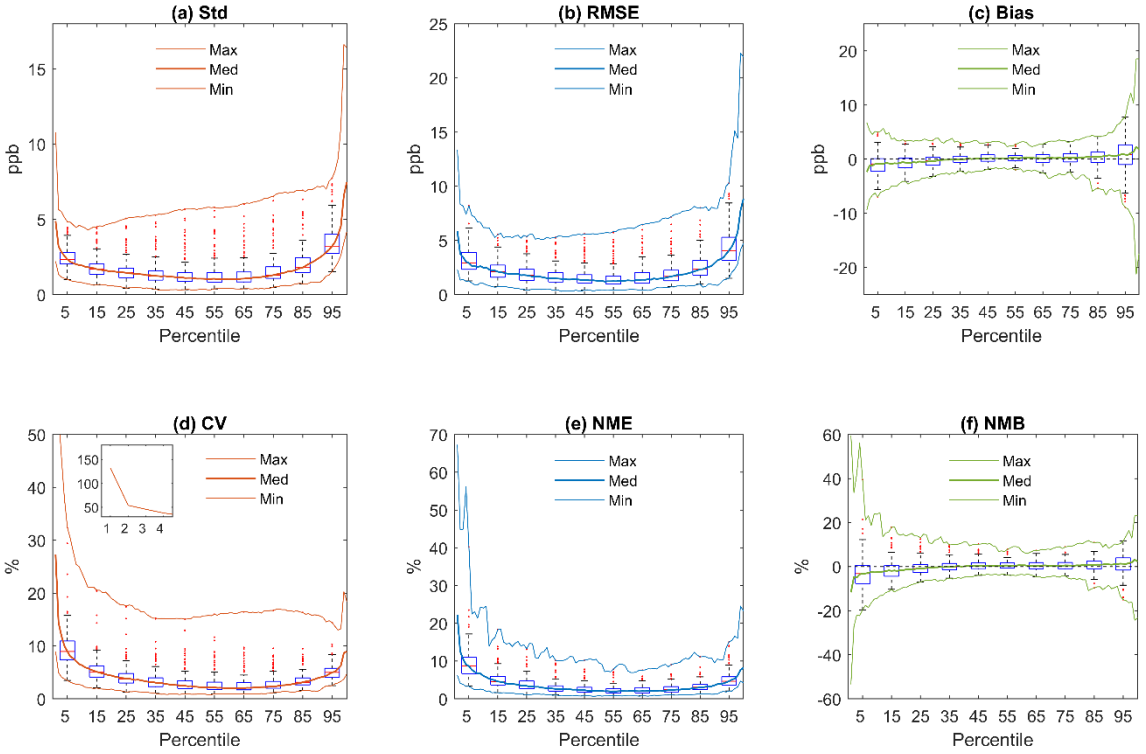


Figure ~~2a~~3a: Comparison between the observed cumulative distribution function (CDF) for 2010 shown in red with 30+ pseudo-observations CDFs generated from historical DM8HR ozone time series shown in ~~light blue~~gray at a suburban site ~~(~~at Altoona in PA (AQS station identifier 420130801~~) at Altoona in PA.).~~ The ~~dark~~ blue line represents the average of the 30+ ~~light blue~~gray lines; Figure ~~2b~~3b: Display of various statistical metrics (standard deviation (std), root mean square error (RMSE), bias) derived by comparing the actual observed and pseudo ozone values in Fig. ~~2a~~3a; Figure ~~2c~~3c: Normalized statistical metrics~~.~~ of normalized mean error (NME), normalized mean bias (NMB), coefficient of variation (CV). Notice the large variability occurring at the lower and upper percentiles.


(a) Std
Max
Med
Min
ppb
Percentile
(b) RMSE
Max
Med
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Percentile
(c) Bias
Max
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ppb
Percentile
(d) CV
Max
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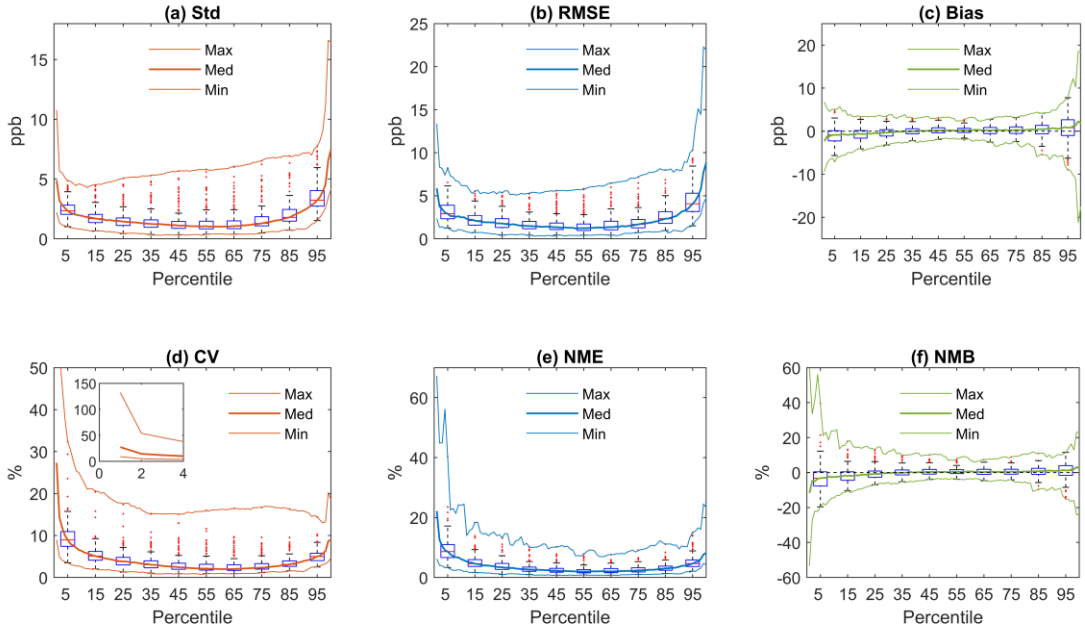
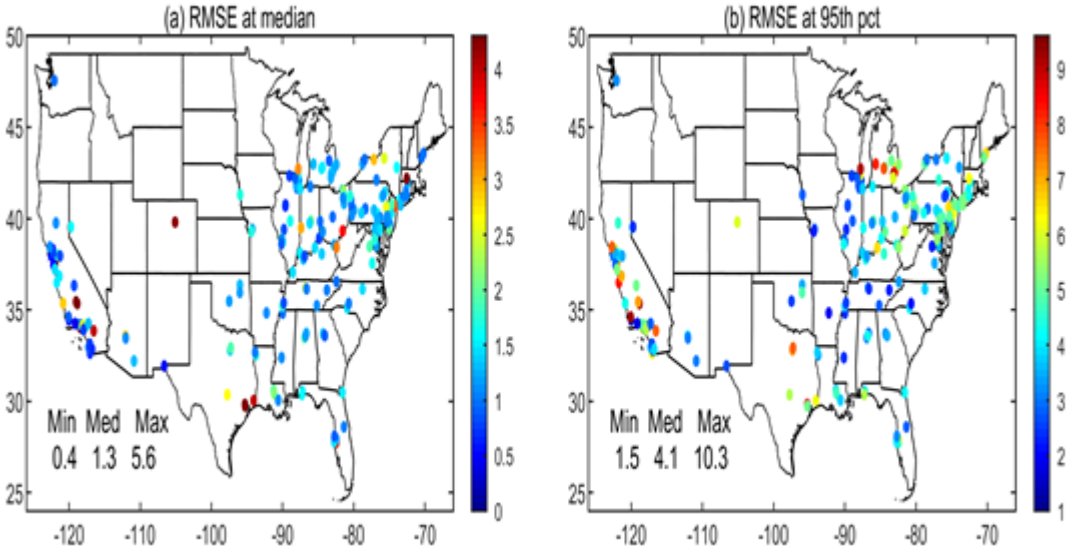
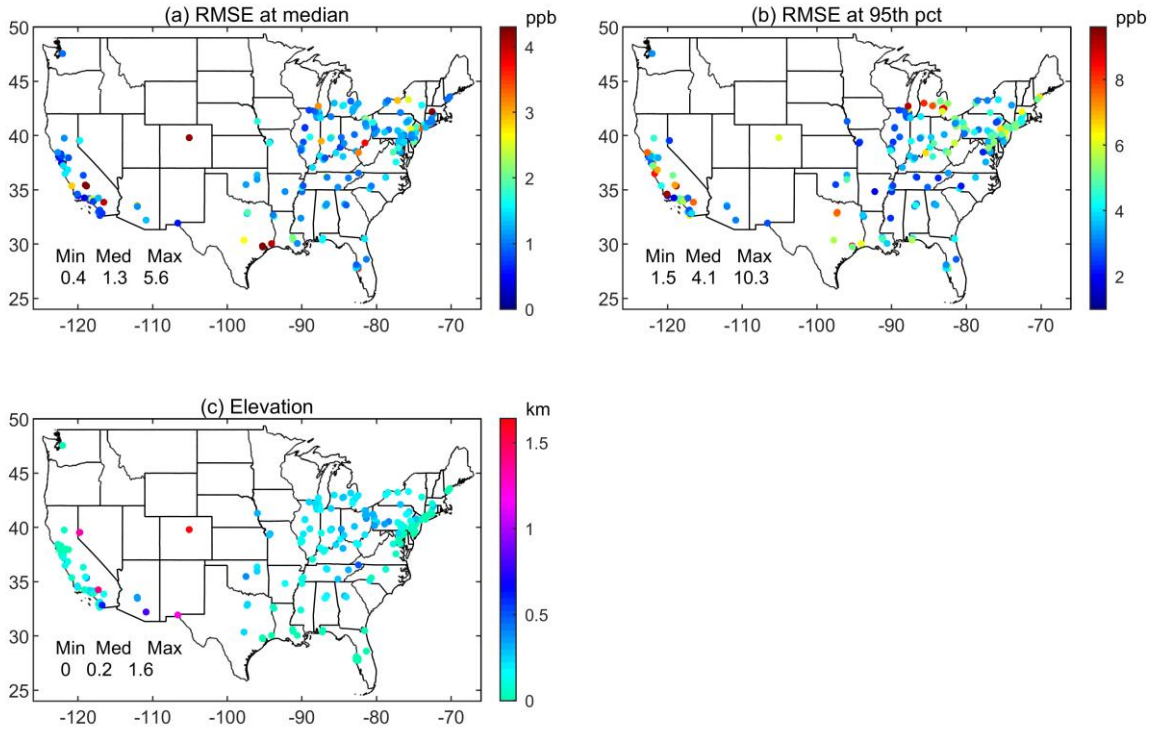


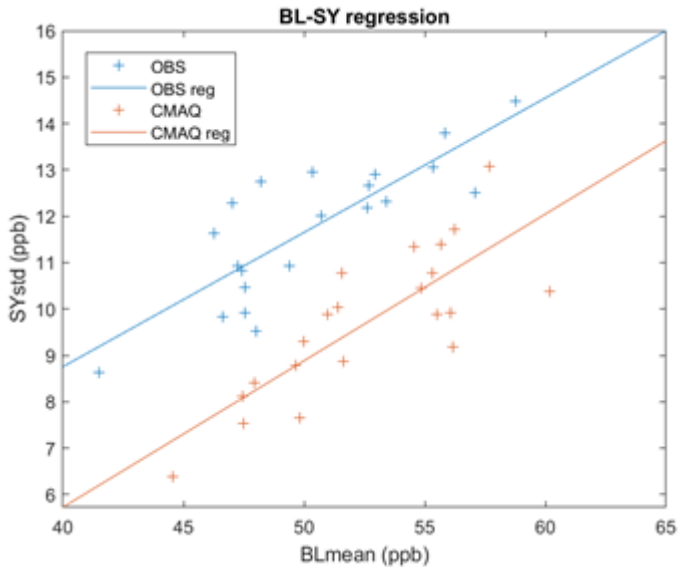
Figure ~~3~~4. Box plots of statistical metrics based on the results from the analysis of DM8HR data at 185 monitoring sites: (a) Standard deviation, (b) Root mean square error, (c) Mean bias, (d) Coefficient of variation, (e) Normalized mean error, and (f) Normalized mean bias. The lower and upper edges of the boxes represent the 25th and 75th percentile values while the whiskers represent the 5th and 95th percentiles. See data analysis procedures using the ozone baseline observed in the year 2010 as the target BL in equations 7 and 8 of Luo et al. (2019).

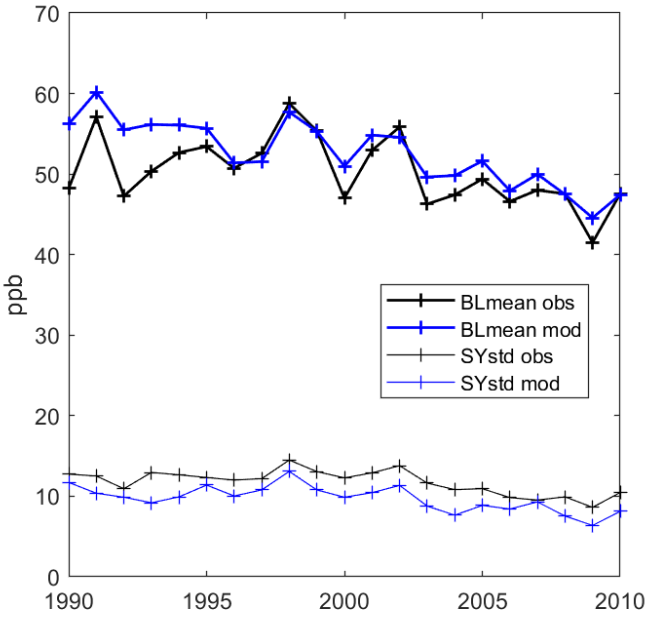
~~. See data analysis procedures using the ozone baseline observed in the year 2010 as the target BL in equations 7 and 8 of Luo et al. (2019).~~


(a) RMSE at median
Min Med Max
0.4 1.3 5.6
(b) RMSE at 95th pct
Min Med Max
1.5 4.1 10.3



~~Figure 4.~~Figure 5. Spatial distribution of the lower bound for the RMSE or expected RMSE at each monitoring site over CONUS (a) at the median and (b) at the 95th percentile; (c) elevation (km) above the mean sea level of each monitoring site.





~~Figure 5. (a) Scatter plot of the standard deviation (i.e., strength) of the SY component vs. the mean of the baseline (BL) component for each of the 21 years from 1990 to 2010 at the Altoona, PA monitoring site.~~

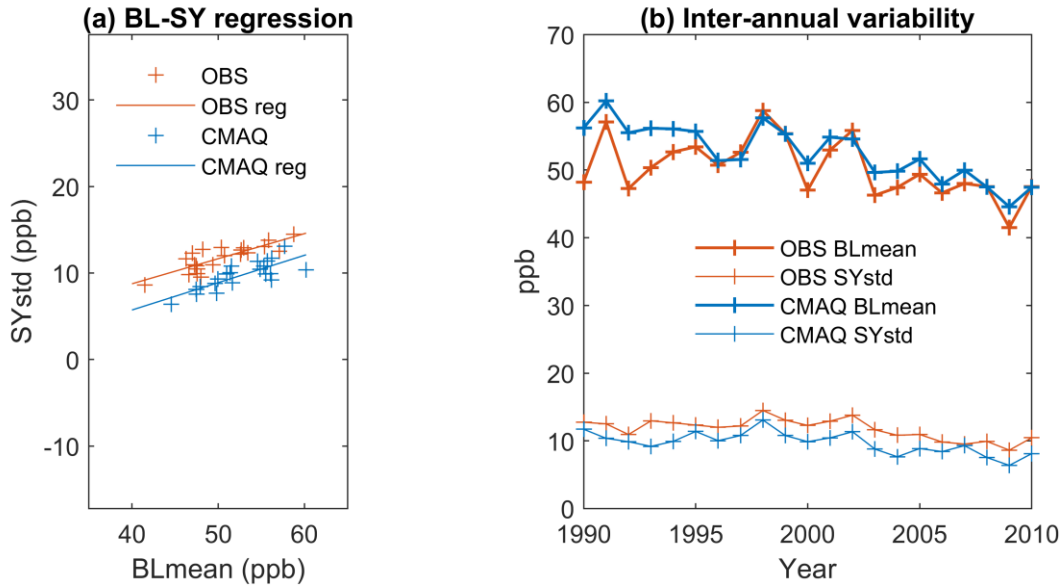
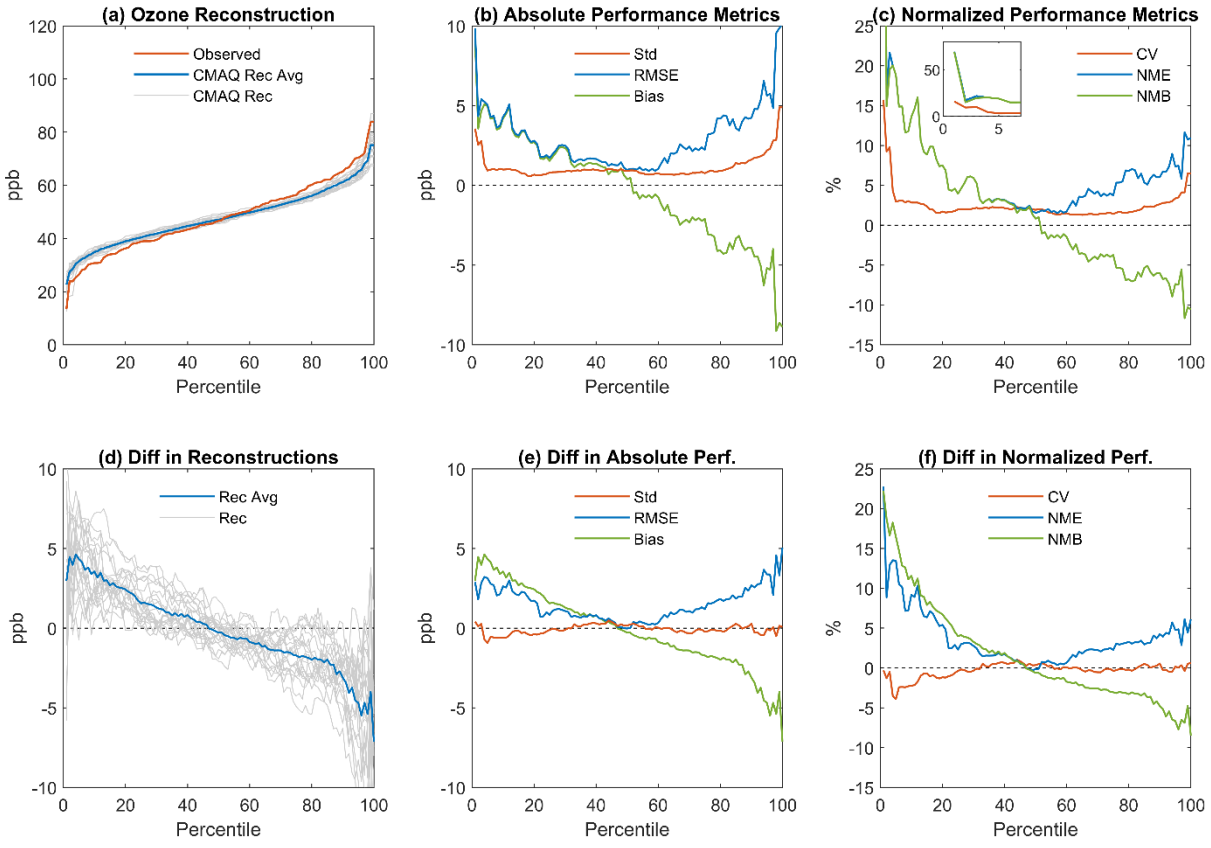


Figure 6. (a) Scatter plot of the standard deviation (i.e., strength) of the SY component vs. the mean of the baseline (BL) component for each of the 21 years from 1990 to 2010 at the Altoona, PA monitoring site. Observations are shown in ~~blue~~red while WRF-CMAQ results are shown in ~~red~~blue. (b) Inter-annual variability in the mean of the baseline component and standard deviation of the synoptic component in the WRF-CMAQ model and observations at the Altoona, PA site. Although year-to-year variation is captured, the model has overestimated the baseline forcing and underestimated the synoptic forcing.


(a) Ozone Reconstruction
Observed
CMAQ Rec Avg
CMAQ Rec
ppb
Percentile
(b) Absolute Performance Metrics
Std
RMSE
Bias
ppb
Percentile
(c) Normalized Performance Metrics
CV
NME
NMB
%
Percentile
(d) Diff in Reconstructions
Rec Avg
Rec
ppb
Percentile
(e) Diff in Absolute Perf.
Std
RMSE
Bias
ppb
Percentile
(f) Diff in Normalized Perf.
CV
NME
NMB
%
Percentile

~~Figure 6.~~

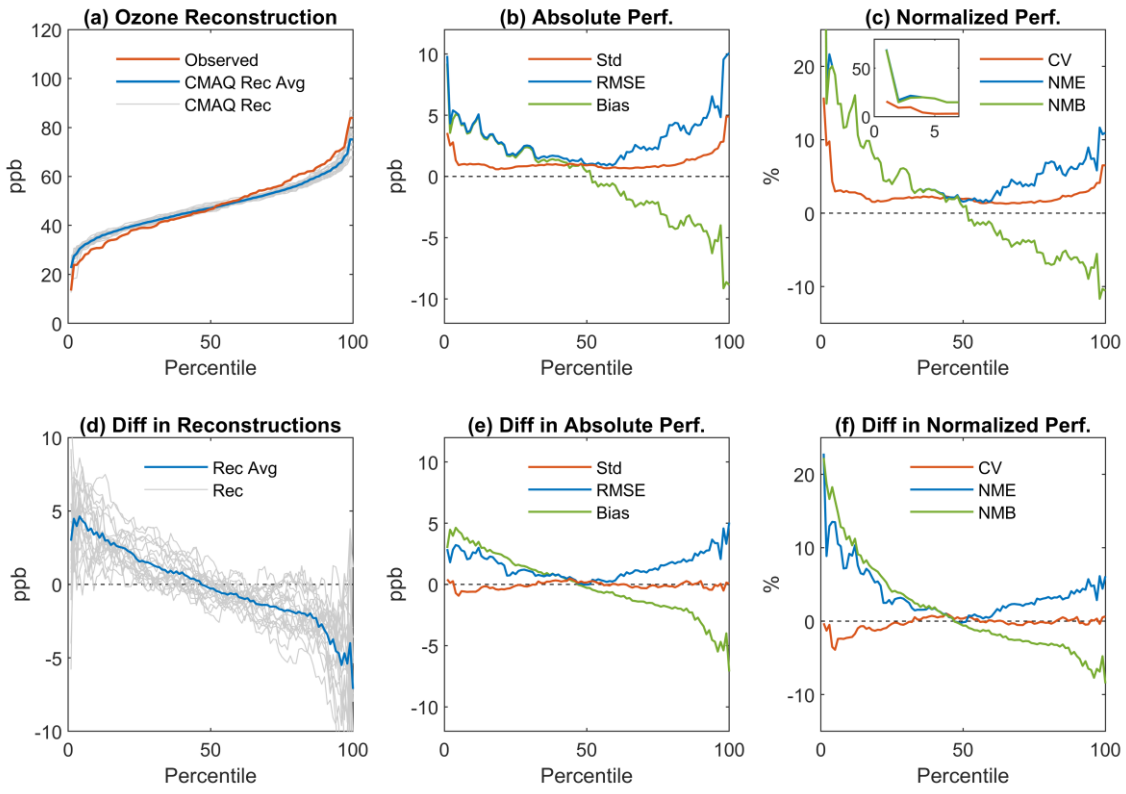


Figure 7. a) Comparison between the observed CDF ~~overlain~~overlaid on 21 'pseudo-simulated' or reconstructed ozone CDFs with SY generated from modeled DM8HR ozone time series at a suburban site ~~(~~420130801) at Altoona in PA~~;~~ (AQS station identifier 420130801); b) Display of various statistical metrics derived by comparing the actual observed and pseudo-simulated ozone values in Fig. ~~6a; 5~~7a; c) Normalized statistical metrics; d).Difference between the pseudo-simulated CDFs shown in Figure ~~6a~~7a and the pseudo-observed CDFs as shown in Figure ~~2a~~7a but calculated from 21 years (1990-2010) of observations only. The ~~light blue~~gray lines represent the differences for a specific SY year while the ~~thick~~ blue line represents the differences between the means of the 21 reconstructions; e) Difference between the absolute performance metrics for pseudo-simulations shown in Figure ~~6b~~7b and those calculated for pseudo-observations as shown in Figure ~~2b~~7b but calculated for 21 years (1990-2010) only. f) As in panel e) but for normalized performance metrics.

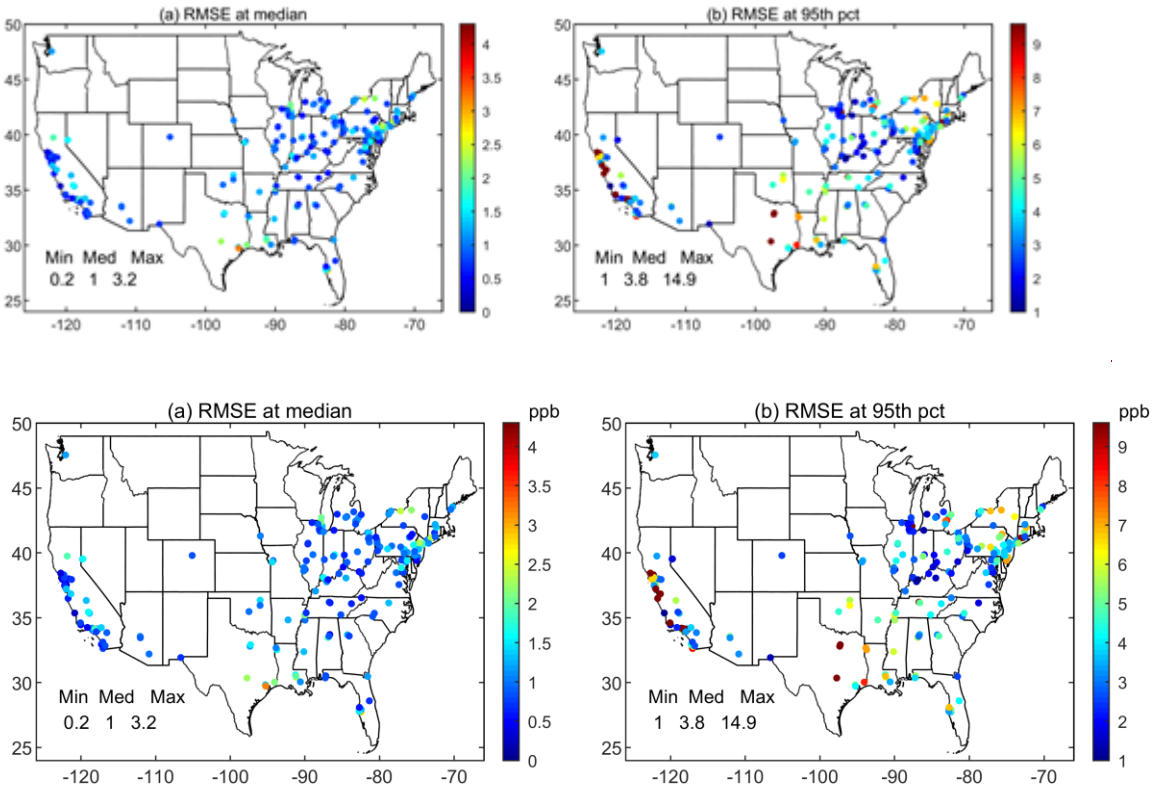


Figure 8. ~~Figure 7.~~ Errors ~~attributable to~~ in the ~~different synoptic forcings in model results~~21 'pseudo-simulated' or reconstructed ozone time series with SY generated from modeled DM8HR ozone time series using BL obtained from observations at (a) the median and (b) 95th percentile.