Response to Reviewer #3's comments on He et al. 2019 Atmospheric Physics and Chemistry manuscript

We thank the anonymous reviewer for thoroughly reading our manuscript and providing helpful comments and suggestions, which lead a significant improvement of our manuscript. The detailed responses to major point comments are listed below (text in *italic* and black is the reviewer's comments, and the normal text highlighted in blue is our response):

This manuscript presents a modeling study of the decadal ozone trend in the US. I am impressed by the significance of the results, but there are still several important issues need to be addressed before publication.

Response: We appreciate the positive comments from the anonymous reviewer, and the manuscript has been revised according to these comments as listed below.
*Line numbers are based on the revised clean version of the manuscript.

1 How will the results of CWRF-CMAQ differ from WRF-CMAQ? You can also use WRF to simulate decadal climate with long-term reanalysis. Better to include some description of the advantage of CWRF over WRF.

Response: The CWRF was developed as a Climate extension of the WRF model incorporating numerous improvements in representation of physical processes and integration of external forcings that are crucial to climate scales, including interactions between land-atmosphere-ocean, convection-microphysics and cloud-aerosol-radiation, and system consistency throughout all process modules (Liang et al., 2012; Qiao and Liang, 2015; Chen et al., 2016; Liu et al., 2016; Qiao and Liang, 2016). It is built with a comprehensive ensemble of many alternate mainstream parameterization schemes for each of key physical processes. To better illustrate the advantage of CWRF, we added this short description in Line 127 of the revised manuscript.

2 Scaling factors were used to get historical emissions. However, this will keep the spatial distribution the same at 2011 level. Why don't you use the information from other historical inventories, such as EDGAR? Could you discuss how this will affect the results?

Response: This is a good question. The NEI2011 inventory adjusted with the ground and satellite measurements provides the best available anthropogenic emissions to the CONUS, which has also been used in the operational U.S. national air quality forecast. We used the U.S. National Emissions Trends to produce the scaling factors to generate historical emissions. It is not a perfect solution as pointed out by the reviewer that the assumption of the same spatial distribution may not be true. However, this method guaranteed that the domain total emissions would be consistent with the U.S. official emission trends, which we believe are the best available emissions for CONUS. We believe it is more important to provide total CONUS emissions constrained by the trend data than emissions with more detailed geographic distributions, when our modeling system has relatively coarse spatial resolution and integrates over 20 years. To explain our approach, we added the following sentences in Line 166 as "*Emissions of the baseline year are based on EPA NEI2011 inventory which can provide the best available anthropogenic emissions to the CONUS and are currently used in the operational U.S. national air quality forecast. The usage of APET scaling factors can guarantee the domain total emissions are consistent with the U.S. EPA emissions trend, although assuming the same spatial* *distribution of anthropogenic emissions from year to year may not be realistic. Without a reasonable observation of actual spatiotemporal variations, it is the cost-effective approach as a first-order approximation to simulate long-term U.S. air quality driven by consistent CONUS total anthropogenic emissions that account interannual trends*".

3 Chemical initial and boundary conditions were obtained from the default concentration profiles built in CMAQ. For long lived chemical species like ozone, long range transport and stratospheric intrusions would be important. If default concentration profiles are set, how to consider the historical changes in sources outside the US?

Response: We understand that the ICs and BCs are important to the CMAQ performance. To reduce the impacts of ICs, we spin-up the CMAQ model for two weeks before the 5-yr continuous simulations (e.g., two weeks in December 1999 were used to create ICs for 2000-2004 CMAQ simulations). Based on our experiences in CMAQ modeling and the EPA guidance, two weeks' spin-up should be able to eliminate the influences from ICs.

The stratospheric intrusions are important for the ozone pollution in high altitude regions such as Denver, Colorado. However, the regional CMAQ does not include stratospheric chemistry, and our model top level is at 50 hPa. A potential vorticity (PV)-O_3 parameterization was developed recently for the hemispheric CMAQ model (Xing et al., 2016), but it is not available for the regional CMAQ version used in this study. We have discussed this shortcoming of our modeling system in the discussion section.

We agree that the long-range transport (LRT) through BCs can play an important role in the regional modeling of U.S. air quality. Our previous studies (He et al., 2016; He et al., 2018) show that the LRT can contribute up to 10% of ozone and $PM_{2.5}$ in the western United States; these numerical simulations are conducted in relatively short period (5 years) under multiple scenarios with fixed and dynamic LBCs. For climate studies, 5-year continuous integration is usually treated as the minimum time period, while longer simulation is preferred to better capture the climate signature. In this project, we designed a 25-yr experiment from 1990 to 2015, so CWRF downscaling can better represent the regional climate of the CONUS. Due to limited computing resources, we chose not conducting 25-yr global CTM simulations to generate dynamic LBCs conditions for CMAQ, but focused on the ozone pollution change within the United States. We understand that our approach could introduce some uncertainties in this study, and added the following sentences in Line 398 of the discussion session, "*So our current modeling system cannot take the historical changes of air pollution outside the United State into account. That is, the effect of long-range transport of air pollutants through model domain boundaries is presumed to be secondary to the long-term trends over the United States."* and In Line 403 *"With these increased air pollutant transported into the United States, our study may underestimate the impacts of domestic emission reductions to U.S. ozone pollution, especially in the West Coast and the Southwest.*".

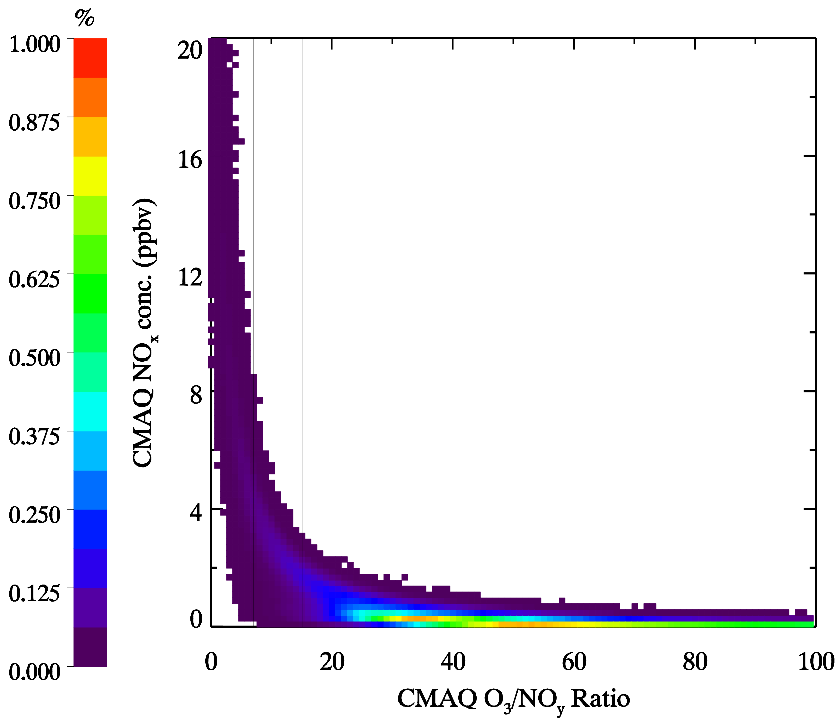
4 O3/NOy ratio was used as the indicator of VOC or NOx limited. The threshold of was adopted (O3/NOy < 15 indicating the VOC-sensitive regime). How to demonstrate this threshold and ratio is proper and accurate or represent the sensitivity. As model usually has difficulty in capturing the concentrations of NOy, the results might be questionable with this assumption.

Response: We appreciate the reviewer raised this concern, which is also pointed out by the other reviewer. First, we understand that computer models have difficulty to accurately capture the ambient NO_y concentrations. In this study, because we did not access long-term research grade NO_y observations from EPA, all the analysis using O_3/NO_y ratios as the photochemical indicators is based purely on CMAQ simulations. So we have to rely on results from the previous studies. Sillman explored the concept using photochemical indicators including O_3/NO_y to identify the regime of ozone photochemical production, finding that the link between the ozone production sensitivity and these indicators is largely unaffected by changes in model assumptions, including emission rates of anthropogenic and biogenic species (Sillman, 1995; Sillman et al., 1997). Observations from urban areas of Atlanta, New York, and Los Angeles was compared with modeling results from the Urban Airshed Model at urban scales, and a threshold of 7 was proposed for using O_3/NO_y ratios as the photochemical indictor (Sillman et al., 1997). Zhang et al. (2009) expanded the study to the CONUS with 1-year CMAQ simulations, suggesting a threshold of 15 for O_3/NO_y ratios. Zhang et al. (2009) used previous CMAQ version 4.4 for this 1-yr CONUS simulations of 2001 at a relatively coarse spatial resolution (36 km), which is close to our 30-km CONUS domain, so we adopted their proposed threshold. We agree that the current manuscript lacked the evaluation of this threshold with our modeling system, and we developed the following approach to test it.

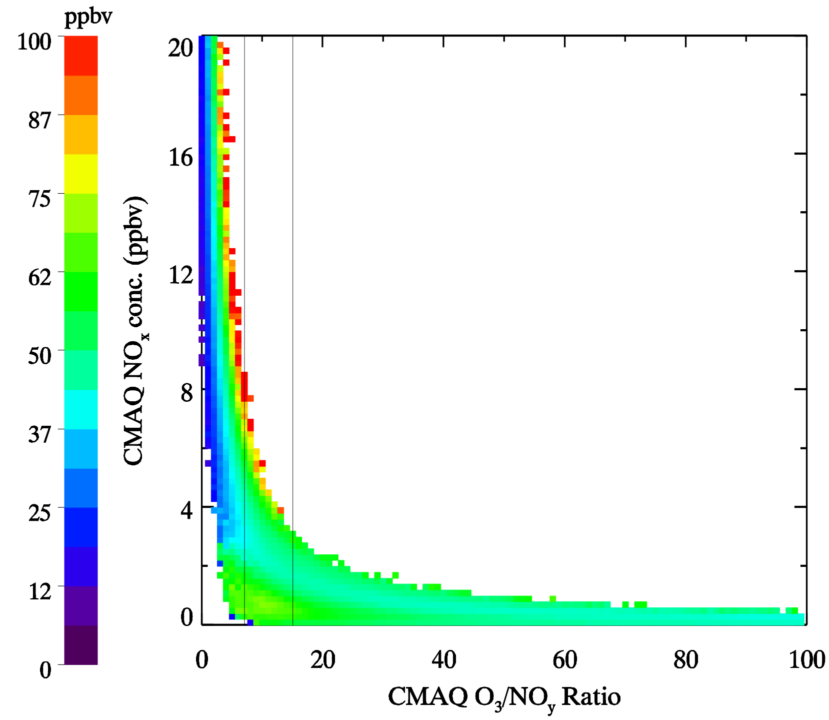
We selected hourly O_3, NO_y, and NO_x concentrations from CMAQ in the afternoon (defined as 12 pm to 4 pm) in 2014, and calculated the O_3/NO_y ratios. Figure 1a shows scatter density of O_3/NO_y ratios vs. NO_x concentrations, which is calculated based on a 100 × 100 bins with NO_x from 0-20 ppbv NO_x (i.e., 0.2 ppbv per bin) and 0-100 O_3/NO_y ratios (i.e., 1 per bin). In the afternoon over the CONUS, the ozone production is mainly in high O_3/NO_y ratio (>15) and low NO_x (less than 2 ppbv) environment, i.e., in the NO_x-sensitive regions by thresholds proposed by both Sillman et al. (1997) and Zhang et al. (2009). Figure 1b shows the same density plot, but the color stands for mean O_3 concentrations. Both low and high ozone concentrations exist in high NO_x region (NO_x > 4 ppbv), which are usually urban or suburban. Then we calculated the weighted ozone concentrations that equals to the product of O_3/NO_y and NO_x scatter density (Fig. 1a) and mean O_3 concentrations (Fig. 1b), which stands for the O_3 sensitivity with respect to O_3/NO_y ratios and NO_y concentrations over the CONUS (Fig. 1c). At the national scale, when the weighted ozone concentrations increase with CMAQ NO_x levels, the photochemical production is NO_x-sensitive. The region with O_3/NO_y higher than 7 and 11 both have this characteristics, while due to low probability (Fig. 1a) and urban environment (Fig. 1b) we believe the O_3/NO_y threshold of 7 stands for the urban environment. Thus, the O_3/NO_y ratio threshold of 15 is more proper for the CONUS scale analysis. This analysis qualitatively supports our application of results from Zhang et al. (2009).

In summary, due to limited resources and experiment design, identifying a threshold of O_3/NO_y ratio is beyond the scope of this study. Using results from our CMAQ model, we proved that the threshold of 15 should be practical for our study. We added the following sentences in Line 323 as "*The usage of O_3/NO_y ratio was first proposed by Sillman (Sillman, 1995; Sillman et al., 1997). Sillman et al. (Sillman et al., 1997) conducted a case study of observations in urban areas (Atlanta, New York, and Los Angeles) and modeling results from the Urban Airshed Model and suggested the threshold of 7 as the transition region from VOC-sensitive environment to NO_x-sensitive environment. Zhang et al. (2009a; 2009b) expanded this method to the CONUS with 1-year observations and CMAQ simulations (36-km spatial resolution) and suggested a threshold of 15 for ozone pollution at the national scale. In this study, we did not have access to the long-term research grade NO_y observations from the AQS network and did not conduct sensitivity experiments (due to computational resource limit) with reduced NO_x emissions following Sillman* *et al. (1997), so we have to reply on the O_3/NO_y threshold from literature. We conducted a simple evaluation of our CMAQ results and found the threshold of 7 could be more proper for urban areas and the threshold of 15 should be more applicable for our study of the whole United State (Figure S1 in the supplementary material). Please note that the O_3/NO_y ratio could depend on the modeling framework, so due to the similarity of our modeling system (30-km CMAQ) and the model used in Zhang et al. (2009a; 2009b), our analysis suggests the similar threshold of 15*" and the discussion above to the supplementary material.

a)



b)


ppbv
100
87
75
62
50
37
25
12
0
CMAQ NO_x conc. (ppbv)
20
16
12
8
4
0
0
20
40
60
80
100
CMAQ O_3/NO_y Ratio

c)

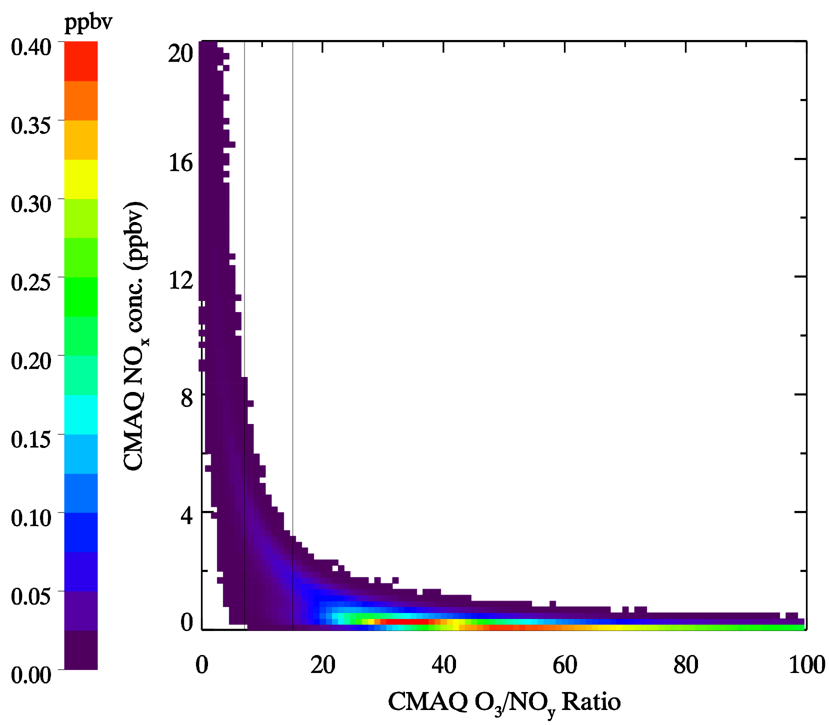

ppbv
0.40
0.35
0.30
0.25
0.20
0.15
0.10
0.05
0.00
CMAQ NO_x conc. (ppbv)
20
16
12
8
4
0
0
20
40
60
80
100
CMAQ O_3/NO_y Ratio

Figure 1. Afternoon O_3/NO_y ratios vs. NO_x concentrations simulated by CMAQ in 2014. a) Scatter density, the color contour stands for the probability for each bin; b) O_3 concentrations, the color contour stands for the mean O_3 over the bins; c) Weighted O_3 concentrations. Two black lines stand for the O_3/NO_y ratios of 7 and 11.

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