### Response to referee comments on "Influence of Synoptic Patterns on Surface Ozone Variability over the Eastern United States from 1980 to 2012"

We thank the referees for their valuable and thoughtful comments, which have improved our manuscript. This document is organized as follows: the Referee's comments are *in italic*, our responses are in plain text, and all the revisions in the manuscript are shown in blue. Boldface blue text denotes text written in direct response to the Referee's comments. The line numbers in this reply refer to the updated manuscript.

### **Referee 1**

**Q1.** The authors investigated the effect of synoptic-scale weather patterns on observed MDA8 surface ozone over the eastern US during summers of 1980 – 2012. They did a fairly interesting study to be quantitative about the effect of the polar jet, the Great Plains low level jet (GPLLJ), and the Bermuda High on surface O3 in the eastern US by examining the relation between the three major EOF patterns and key meteorological variables followed by correlation between seasonal mean MDA8 ozone and variables representing those three synoptic systems. They constructed a model to predict seasonal MDA8 ozone averaged in four regions of the eastern US.

Response. We have revised the text to more strongly convey a few other main points.

(1) At the end of the Abstract, we now say:

P2, L12-16. Our work underscores the impact of synoptic patterns on ozone variability and suggests that a combination of changing local and synoptic meteorology together with trends in background ozone will influence U.S. ozone air quality in future decades. The observed relationships of U.S. surface ozone and synoptic circulations in this study can also be used to validate models of atmospheric chemistry.

(2) In the Discussion and Conclusion section we have added the following text:

P21, L12-15. None of the three identified synoptic circulations show significant trends in the 1980-2012 timeframe. Our result supports the conclusion in Cooper et al. (2012) that the observed decreasing ozone trend is mainly caused by emission control.

(3) We have also added a new paragraph in the Discussion and Conclusion.

P23, L1-17. Our work identifies the synoptic patterns that strongly influence the variability of U.S. surface ozone, and it provides a set of metrics that may be used to evaluate the skill of chemical transport models (CTMs) and chemistry-climate models (CCMs) in capturing this influence. Few chemistry-climate studies to date have documented either the model capability in capturing the synoptic patterns important to ozone or the sensitivity of modeled ozone to these patterns. For example, using the GFDL-AM3 model, Rasmussen et al. (2012) evaluated only the relationship of ozone with local temperature and not with synoptic patterns. Turner et al. (2013), however, found that this model underestimates the dependence

of ozone in the northeast United States on cyclone frequency. To our knowledge, no model study has examined the effect of the westward extent of the Bermuda High on calculated levels of ozone in the southeast United States. As noted by Fiore et al. (2009) and Parrish et al. (2014), CTMs and CCMs have difficulty in simulating observed ozone variability on both seasonal and multi-year timescales, and at least part of this difficulty may be due to model deficiencies in the representation of synoptic patterns and their impact on surface ozone. By testing the sensitivity of modeled ozone to the synoptic patterns we identify here, a clearer picture of the causes of model discrepancies should emerge.

**Q2.** The time period of the data set used in the study needs to be clarified up front. The authors stated that they were focused on the latest 20 years (1993-2012) at one point (line 2, page 13079). A few lines down they stated that they used data in the eastern US over 1980-2012 for the EOF analysis. When they examined the relation between the polar jet and surface ozone concentrations in Section 5, they used 1980-2012 (lines 23, page 13083).

**Response**: We have updated our approach. We now use all available data up to 2012 in both EPA-AQS and CASTNET for our analyses, expanding the number of sites in both datasets and the time frame for CASTNET.

P6, L19-21. We include all sites with at least one summer of observations, resulting in a network of 1670 sites for AQS over 1980-2012 and 72 sites for CASTNET over 1990-2012 in the eastern United States.

In addition, we have revised the text and Figure 1 to make clear why the observed bimodal structure in ozone variability is robust across all time spans and site types.



**Figure 1**. Latitudinal variation of zonal mean ozone standard deviation (a, c) and relative standard deviation (b, d) from CASTNET (a, b) and AQS (c, d), averaged between 100°W and 65°W longitude and binned to 2° intervals in latitude. All values are for JJA mean MDA8 ozone. The different colors denote different 5-year time intervals, except for the most recent interval, which is 8 years in length.

P6, L22.

Figure 1 reveals a north-south bimodal structure in the zonally averaged variability of daily surface ozone across the eastern United States for a range of timespans. To calculate ozone variability, we proceed as follows. First, we calculate the ozone SD and relative daily SD for each site in each summer. The relative SD is obtained by dividing the ozone SD by the mean JJA MDA8 ozone for that summer. Second, we average both kinds of ozone SD over each 2° latitude bin across the eastern United States for each time span. By first calculating daily SD for each summer, and then taking the average across summers, we remove much of the influence of changing NOx emissions during this time interval. The relative ozone SD further isolates the effect of meteorology by normalizing with mean JJA ozone.

As shown in Fig. 1, both the absolute SD and the relative SD exhibit a bimodal structure over a range of time spans in the AQS and CASTNET datasets. Peaks in absolute SD appear between 29°-35°N and 39°-43°N, while peaks in the relative SD appear between 28°-32°N and 40°-45°N. From 1980 to 2012, the magnitudes of both kinds of SD diminish due to reductions in the emissions of ozone precursors. The northern peak of SD decreases more rapidly than the southern one (Fig. 1a and 1c), and this result can be explained by the more dramatic NO<sub>x</sub> decreases in the North (Russell et al. (2012)). These results are consistent with Bloomer et al. (2009), who found that the ozone-temperature slopes decreased in all ozone percentiles after implementation of the 2002 NO<sub>x</sub> controls, and that these decreases were greatest for ozone in the higher percentiles. Our finding that the relative SD declines over time (Fig. 1b and 1d) is also consistent with Bloomer et al. (2012), as high levels of ozone drop more rapidly than does mean ozone. We also examine the sensitivity of the bimodal structure to the AQS site types (rural, suburban and urban) and find that the relative SD shows a clear bimodal structure for all time spans and site types (Fig. S1). The trend in emissions changes only the magnitude of relative SD, with decreases at all latitudes, but it does not erase the bimodal structure. Our results suggest that the ozone relative SD provides a useful metric to gauge the influence of meteorological variability on ozone even as anthropogenic emissions change over time. The persistence of the bimodal structure in relative SD throughout the time period at all AQS site types and at all CASTNET sites increases our confidence that these peaks signify the influence of meteorology and not that of high emissions of anthropogenic precursors.

**Q3**. How many AQS sites are urban and rural? Did the authors separate them and find results to be any different from all used? The bi-modal pattern was more pronounced in the CASTNET data than in the AQS data. If the urban data were removed from the AQS data set, how would the pattern of latitudinal variation look using the AQS data? Major urban areas are along the periphery of the eastern US, so would the greatest relative SD still occur there after urban data were removed?

**Response**. The reviewer makes an excellent suggestion. In a new figure in the Supplement, we now show results classified by type of EPA-AQS site. As described below, the bimodal structure in relative SD occurs at all types of sites.



**Figure S1**. Latitudinal variation of zonal mean ozone concentrations (a, d, g), ozone standard deviation (b, e, h), and relative standard deviation (c, f, i). All values are for JJA mean MDA8 ozone. The top row shows results from rural sites, the middle from suburban sites, and the bottom from urban sites in AQS. Data have been averaged between  $100^{\circ}W$  and  $65^{\circ}W$  longitude and binned to  $2^{\circ}$  intervals in latitude. The different colors denote different 5-year time intervals, except for the most recent interval, which is 8 years in length.

#### **Supplement P1-2**

## 1. Spatial patterns of temporal variability in daily JJA surface ozone in the eastern United States in different site types

Here we examine whether the bimodal structure of observed ozone variability is sensitive to the type of EPA-AQS site (rural, suburban, and urban). Fig. S1 shows the zonally averaged ozone concentrations, standard deviation (SD), and relative SD for the three site types across the eastern United States for different time intervals. As in Fig. 1, we include in our analysis all sites reporting at least one observation every summer during 1980-2012, resulting in 787 rural, 617 suburban, and 266 urban sites altogether. Zonally averaged ozone concentrations at all site types reveal a unimodal structure peaking around 35°N-40°N for all time spans, with an overall trend of decreasing concentrations from 1980 to present. The absolute ozone SD shows a bimodal structure for all time spans at rural sites, with peaks at 29°-35°N and 39°-43°N. At urban and suburban sites, the northern peak in absolute SD diminishes over time, while the southern peak persists, consistent with the more rapid NOx reductions in the North (Russell et al., 2012). In contrast, the relative ozone SD shows a clear bimodal

structure for all three site types and all time intervals. In calculating the relative SD, we remove much of the influence of anthropogenic emissions. Our finding that the bimodal structure is a robust feature in both the absolute and relative ozone SD at rural sites thus reflects the smaller influence of anthropogenic emissions at such sites. The persistent bimodal structure of the relative SD in ozone across time and sites therefore gives us confidence that the relative SD can be used to identify those regions where meteorology plays a large role in driving ozone variability.

**Q4.** It is hasty, without support evidence, to decide that larger ozone SD was a result of changes in synoptic meteorology, not anthropogenic emissions, when one looks at a 20 year dataset (Lines 23-26, page 13078; lines 1-2 on page 13080). If a season of data was examined, this statement would likely be valid. As we are all aware the annual 98th percentile mixing ratio of NOx has decreased by 46% in national average from 1990 to 2013, suggesting great reductions in NOx emissions. Over this time span, ozone SD could be fairly large due to emission reductions of this magnitude alone. In lines 24-29 on page 13079, the authors stated that a 2 ppbv discrepancy in ozone SD between the CASTNET and AQS data were likely due to the inclusion of many urban sites in the latter. If this was true, it would mean that the influence of anthropogenic emissions ozone SD was not trivial, which appears to contradict what they decided early on. Again, it seems necessary to remove the urban data from the AQS dataset to see if results would be different.

**Response**. The reviewer makes a good point about the potential influence of anthropogenic emissions on ozone SD at urban sites, and we have extended our analysis of ozone variability, as detailed in our responses to Q3.

P7, L21-24. The persistence of the bimodal structure in relative SD throughout the time period at all AQS site types and at all CASTNET sites increases our confidence that these peaks signify the influence of meteorology and not that of high emissions of anthropogenic precursors

Supplement P1 L21-24. The persistence of the bimodal structure in relative SD throughout the time period at all AQS site types and at all CASTNET sites increases our confidence that these peaks signify the influence of meteorology and not that of high emissions of anthropogenic precursors.

We have also clarified our approach to calculating SD. Our approach removes the potential problem identified by the Reviewer in calculating SD over long timespans.

P6, L23-30. To calculate ozone variability, we proceed as follows. First, we calculate the ozone SD and relative daily SD for each site in each summer. The relative SD is obtained by dividing the ozone SD by the mean JJA MDA8 ozone for that summer. Second, we average both kinds of ozone SD over each 2° latitude bin across the eastern United States for each time span. By first calculating daily SD for each summer, and then taking the average across summers, we remove much of the influence of changing NOx emissions during this time interval. The relative ozone SD further isolates the effect of meteorology by normalizing with mean JJA ozone.

**Q5.** The EOF analysis suggested 24%, 18%, and 11% of the total variance of surface DMA8 ozone data can be explained the first three EOF patterns, which were hypothesized to be linked to the polar jet, Bermuda High, and the GPLLJ, respectively. Further the authors studied the relation between the JJA mean DMA8 data and variables representing the three systems and found good correlation. It should be noted that when they did this part of analysis, they used seasonal means. This means that the 53% of the total variance was in fact the portion on seasonal scales. Consequently the model they constructed (Eq. 1) was capable of predicting seasonal averaged MDA8 ozone mixing ratios. Therefore, it is an overstatement that using a single metric of the synoptic systems identified in the study they could predict ozone variability in future climate regimes. I think they might want to be more specific about the time scales and uncertainty of such prediction.

#### **Response:**

Our response to Q5 is in two parts. First, we clarify why we perform the EOF using daily ozone data and then choose to further diagnose ozone variability on seasonal scales. We have also added a new section (Section 3) in the Supplement to compare the EOF results using daily data and monthly data. The second part of our response begins at the bottom of page 9.

#### P8, L23-25.

As we shall see below, the use of daily ozone in this EOF analysis provides a clearer picture of the synoptic scale meteorological variables contributing to ozone variability.

#### P11, L3-L13.

As a test of our approach, we repeat the EOF analysis using mean JJA MDA8 ozone fields, instead of daily mean fields. The top three EOF patterns for the 1980-2012 time period using seasonal mean ozone are similar to those we derive using daily mean ozone (Fig. S3). However the link between EOF3 and 850 hPa geopotential height in this analysis is much weaker than what we find using daily ozone data (Fig. S3i vs. Fig. 3f), and thus the analysis fails to identify the influence of the Bermuda High on ozone air quality (Fig. S3). As we discuss in Sect. 6, the failure arises because the influence of the Bermuda High on ozone varies nonlinearly with the location of the Bermuda High west edge, and the monthly mean EOF analysis obscures this nonlinear relationship. However, the daily dataset with its abundance of observations can more easily reveal this fine structure in the EOF analysis.

#### P11, L14.

In the following sections, we take advantage of information gleaned from the EOF analysis to develop a set of metrics that quantify the relationships between surface ozone and synoptic patterns. Such relationships can be implemented in a simple model and readily applied to archived meteorological output from climate models. Although we have so far focused on daily ozone data, we now turn to seasonal mean ozone data for two reasons. First, the patterns of many synoptic circulations are noisy on daily timescales, making it challenging to define the metrics needed for our simple model. Such circulations include cyclone frequency (Leipensperger et al., 2008), jet wind latitude (Barnes and Fiore, 2013), and the Bermuda

High west edge (Li et al., 2011, 2012). The windspeed of the GPLLJ is easier to characterize on daily timescales, and we discuss the effects of daily GPLLJ windspeeds and surface ozone in the Supplement (Fig. S2). Second, meteorological output from climate models is more widely available on monthly timescales than on daily timescales. Thus our simple model, if it depends on monthly mean meteorology, will be of greater utility to the community.



**Figure S3.** The top panels show (a) the spatial loadings of the first EOF pattern (EOF1) in JJA mean surface MDA8 ozone over the United States from 1980 to 2012 and the correlations *r* between the principal components time series for the first mode (PC1) and (b) JJA mean 500 hPa zonal wind speeds and (c) 500 hPa geopotential height. Composite 500 hPa wind anomalies associated with positive loadings of the first EOF pattern are shown as black arrows in Panel (c). Panel (d) is same as (a), but for the second EOF pattern (EOF2). Also shown are the correlations between PC2 and (e) JJA mean 850 hPa wind anomalies with positive PC2 are shown as black arrows in Panel (f). Panel (g) is same as (a), but for the third EOF pattern (EOF3). Correlations are shown between PC3 and (h) 850 hPa meridional wind speeds and (i) 850 hPa geopotential heights. Panel (i) also shows the composite 850 hPa wind anomalies associated with positive PC3 (black arrows). White areas in all panels indicate either missing data or grid boxes where the correlation is not significant at the 0.05 level.

#### **Supplement P3-5**

## **3.** Use of empirical orthogonal functions to diagnose drivers of seasonal JJA ozone variability

We repeat the EOF analysis using the timeseries of 1980-2012 JJA mean ozone fields, instead of daily mean fields. In using daily data, the first three EOFs explain just 53% of the total variance in ozone; here we find that using seasonal mean data can explain 75% of the total variance. This is because the averaging process from daily to seasonal data can smooth many noises. Figure S3 shows the results of this EOF analysis and the correlations of the PCs with different meteorological variables. The top three EOF patterns are similar to those we derive using daily mean ozone (Fig. 3), but with one slight difference, as detailed below. We first describe the similarities between the two EOF analyses. As in the analysis with daily ozone data, EOF1 here is linked to the southward movement of the polar jet (Fig. S3a-3c) and explains 48% of the total variance. EOF2, explaining 15% of the total variance, exhibits an east-west pattern characteristic of a westward extension of Bermuda High and an enhanced GPLLJ (Fig. S3d-3f). EOF3, explaining 11% of the total variance, displays a northeastsouthwest pattern associated with enhanced 850 hPa meridional wind from the Gulf of Mexico (Fig. S3g and 3h). However, the link between EOF3 and 850 hPa geopotential height is much weaker than what we find using daily ozone data, as can be seen in comparing Fig. S3i and Fig. 3f. This weak relationship makes clear that the analysis using seasonal ozone data fails to identify the influence of Bermuda High west edge on ozone air quality. As we discuss in Sect. 6, the failure arises because the influence of the Bermuda High on ozone varies nonlinearly with the location of the Bermuda High west edge, and the monthly mean EOF analysis obscures this nonlinear relationship. Because the daily dataset provides a greater abundance of observations, the EOF analysis using these data can identify the fine structure of ozone variability.

Also, we have revised and added more description in the main text.

#### P9, L24.

The correlation changes from negative in the Midwest and Texas to positive in the eastern United States, with the change in sign co-located with the strongest correlations of PC2 and meridional transport. The composite wind anomaly associated with the positive PC2 reveals strong onshore winds from the Gulf of Mexico, bringing clean maritime air into the southern Great Plains. As these winds move northward, they likely carry aged polluted air from the Midwest to the Northeast. This mechanism, also identified by Fiore et al. (2003), accounts for the northeast-southwest EOF pattern in Fig. 3d. The 850 hPa geopotential height is sometimes used to diagnose the Bermuda High (e.g., Li et al., 2011). The pattern of wind anomalies in Fig. 3f implies that surface ozone in the deep South is lowest when the western boundary lies at ~85°W longitude and that either the westward or eastward shifts in this boundary increase ozone in this region.

Second, we address the reviewer's comments that "53% of the total variance was in fact the portion on seasonal scales" and that "it is an overstatement that using a single metric of the synoptic systems identified in the study they could predict ozone variability in future climate regimes."

We are not quite sure what the reviewer's intent is here. The number 53% refers to the explained fraction of daily spatial variability in ozone concentrations. The daily EOF analysis provides guidance for us to identify the most important synoptic circulation patterns. These synoptic patterns may explain more temporal variance in ozone at the seasonal scale.

#### P11, L14.

In the following sections, we take advantage of information gleaned from the EOF analysis to develop a set of metrics that quantify the relationships between surface ozone and synoptic patterns. Such relationships can be implemented in a simple model and readily applied to archive meteorological output from climate models.

Also, as pointed out that in the last paragraph of the manuscript, trends in the synoptic circulation patterns we identify contribute only one part of the effect of climate change.

P23, L19-L24. However, local meteorological conditions (e.g., Bloomer et al., 2007) and background ozone levels (e.g., Fiore et al., 2002; Wu et al., 2008; Wang et al., 2009) also influence ozone variability in the East, and future climate change could alter these other factors as well. A complete picture of ozone air quality in the coming decades in this region thus requires consideration of all three factors and quantification of their percent contributions to potential change.

**Q6**. Section 8 is superfluous. It mostly repeats what was already stated in the preceding sections with very little discussion. I suggest that this section be shortened, including only key points and their important implications.

**Response**: We now do more to emphasize the importance of Section 8 to rest of the paper. Following the reviewer's suggestion, we have also shortened the text.

First, we refer the reviewer to the questions posed at the end of the Introduction. P5, L5-8

(1) What is the percent contribution of synoptic meteorology (vs. local meteorology) to the observed variability in U.S. surface ozone?

(2) Have trends in synoptic meteorology either offset or enhanced the ozone air quality gains from emission reductions over the eastern United States in recent decades?

Below is the revised text for Section 8, which addresses these questions. We show this Section in its entirety.

#### P19, L1.

# 8 Percent variability in mean seasonal JJA MDA8 ozone explained by synoptic patterns and trend analysis.

In this section, we quantify the percentage of interannual variability in mean JJA MDA8 ozone that can be explained by the combination of the synoptic patterns identified in this study. We look in particular to see where surface ozone is best explained by these patterns. This step is essential to demonstrate (1) the importance of synoptic scale meteorology to

surface ozone and (2) the need to test the sensitivity of modelled ozone to changing meteorological patterns, especially in chemistry-climate studies. We also examine the observed meteorological data for trends in the key synoptic patterns. Previous studies have reported sometimes contradictory results regarding such trends (e.g., Leibensperger et al., 2008; Turner et al., 2013). Improved knowledge of existing trends in synoptic scale patterns will allow us to assess to what degree the observed decline in U.S. surface ozone can be attributed to cuts in precursor emissions or to meteorology. If trends in synoptic patterns important to ozone levels have indeed occurred and can be traced to ongoing climate change, such information would benefit policymakers as they plan ahead for future air quality.

We first construct a multiple linear regression model to correlate mean JJA MDA8 ozone and three indices of synoptic-scale circulation: the JJA polar jet frequency and the mean JJA longitude and latitude of the Bermuda High west edge. The model is of the form

$$ozone = Jet-freq + 1555-gpm BH-Lon \times I(west) + 1555-gpm BH-Lat \times I(west) + 1565-gpm BH-Lon \times I(east) + 1565-gpm BH-Lat \times I(east)$$
(1)

where *ozone* refers to mean JJA MDA8 ozone in ppbv for each grid box, *Jet-freq* is the JJA jet frequency in the Midwest and Northeast in count grid<sup>-1</sup> summer<sup>-1</sup>, *I(West)* and *I(East)* are indicators of West Regime and East Regime based on the Bermuda High west edge, 1555-gpm and 1565-gpm are the isolines used to calculate Bermuda High west edge, and *BH-Lon* and *BH-Lat* are the mean JJA longitudes and latitudes of the Bermuda High west edge as determined for the relevant regime, with both longitudes West and latitudes North assigned positive values. We use a stepwise procedure to delete terms in Eqs. (1) based on the Akaike Information Criterion (AIC) (Venables and Ripley, 2003). Figure 8a compares the model predictions for mean JJA MDA8 ozone with observations over the 1980-2012 time period across the eastern United States. We find the greatest coefficients of determination ( $R^2$ ) in the Midwest and parts of the Northeast and Southeast, where the polar jet and the Bermuda High west edge together explain 50-80% of the interannual variability of mean JJA MDA8 ozone. Timeseries of observed and predicted mean JJA MDA8 ozone reveal that these two synoptic patterns explain 71% of the total variance in the Midwest, 59% in the Northeast, 53% in the South Central and 59% in the Southeast (Fig. 8b-8d).

We find no significant trend of the identified synoptic patterns over the eastern United States for the 1980-2012 period (Table 2). Our results thus support the conclusion of Cooper et al. (2012) that the observed decrease in afternoon surface ozone from 1990 to 2010 was likely caused by tightening emission controls and not by trends in meteorology. Our work is consistent with Bloomer et al.(2010) who found that surface ozone at five CASTNET sites in the eastern United States declined from 1989 to 2007, despite a warming trend (~0.5 °C decade<sup>-1</sup>) in daytime temperatures. Consistent with the Leibensperger et al. (2008) trend analysis of JJA cyclones crossing Canada, we find a significant decrease of the polar jet frequency (-0.099 count a<sup>-1</sup>, p < 0.1) and polar jet wind speed (-0.067 m s<sup>-1</sup> a<sup>-1</sup>, p < 0.05) over the 1980-2006 time period. However, we find no significant trend in these variables over the extended time period of 1980-2012. Turner et al. (2013) reported more frequent midlatitude cyclones crossing the eastern United States since 2006; consistent with that result, we find increased polar jet frequency since then. Reasons for these short-term variations in polar jet indices are unknown.

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