Response to Reviews

We thank the reviewer for constructive comments to improve the manuscript. The comments are reproduced below with our responses in blue. The corresponding changes in the manuscript are highlighted in blue.

Reviewer #2

While I like the research topic and design of this paper very much and think that it's long overdue for a comprehensive paper on understanding the effects of drought on air quality, some of the main statements are too broad. Several related issues were mentioned in the initial review. If changes were made, it would have made this review simpler.

The reviewer made a comment in his/her initial review about using different cloud fractions (CF) for ozone and aerosols, which we chose to address here in the final revision stage. We agree with the reviewer that total cloud fraction should be used for the analysis of the radiation effects on ozone chemistry, while boundary layer cloud fraction should be used for the analysis of in-cloud oxidation of SO_2. In the revised Figure 8 and Table S2, we have added both total cloud fraction (integrated between 1000 and 10 hPa) and boundary layer cloud fraction (integrated between 1000 and 800 hPa) from the ISCCP observation and the two models (GDFL and GISS) that archived layer specific cloud fractions. The ISCCP data show a 9-24% decrease in total CF and 6-13% decrease in boundary layer CF during drought periods (Table S2). Both the GFDL and GISS model show much larger (30-47%) decreases of total and boundary layer CF. The correlation slope between SPEI and total/boundary layer CF is about 10 times higher in the two models than that from observations (Figure 8). This confirms our original finding that the models tend to underestimate cloud fractions (both total CF and boundary CF) during drought, leading to excessive reductions of in-cloud formation of sulfate aerosols.

I will use the abstract for illustrations. A statement such as "These enhancements show little sensitivity to the decreasing trend of US anthropogenic emissions: : :" is too strong. The decrease of sulfate has been very significant in the US. It is hard to believe that this decrease has no effect on an analysis of the effects of drought on sulfate (or PM2.5 in general). In this paper, there is no comparison of the general decreasing trend of sulfate due to emission reduction with the effects of drought to support the statement.

We agree with the reviewer that a few statements need to be revised to improve clarity and specify scope. The statement in question here does not refer to the actual concentrations of ozone or $PM_{2.5}$, which indeed show a large decrease over 1990-2014 with decreasing US anthropogenic emissions (see Table 2, last column). We meant to say that the pollutant enhancements associated with droughts do not change at the same rate or even the same direction of decreasing anthropogenic emissions in the US. We've revised the statement as: "The pollutant enhancements associated with droughts do not appear to be affected by the decreasing trend of US anthropogenic emissions, indicating natural processes as the primary cause".

Another statement in the abstract "Most climate-chemistry models are not able to reproduce the observed responses of ozone and PM2.5 to drought severity, suggesting a lack of mechanistic understanding of drought effects on atmospheric composition." The results from this paper show that there are deficiencies in climate-chemistry models. These deficiencies do not necessary imply that there are missing mechanisms in the models. In the discussion section of the paper (which I like better than the abstract), uncertainties in the models were described. It seems to me that the model deficiencies are more a problem of model representation of drought events not that there is clear evidence for missing climate-chemistry mechanisms.
Agreed. We've removed the second part of the sentence (i.e. removed "suggesting a lack of …"). Regarding the models' ability to simulate drought, we showed in Figure S7 (Supplementary Material) that the four ACCMIP models were able to capture the observed spatial patterns of drought occurrence frequency. Severe drought (model SPEI < -1.3) occurs ~20% of the time over the west and southern US, consistent with observed SPEI. However, the temporal correspondence (i.e. month-to-month) between model SPEI and global SPEI dataset is weak, largely due to the models deficiency in simulating temporal variability of precipitation. This weak correlation however is not expected to affect the model evaluation, because we used the model SPEI to derive the simulated SPEI-pollutants relationships from each model and compared those relationships between model and observations, rather than ozone or PM2.5 concentrations per se. We've added discussion of the model ability to simulate drought in the manuscript (pg 9, line 33-39).

The statement "Drought thus poses another aspect of climate change penalty on air quality not recognized before" would imply that there were no studies of the kind before. Wang and Xie et al. (2015), for example, obviously discussed some of the issues.
Agreed. In fact, the work of Wang and Xie et al. (2015) referred by the reviewer is our own. The part "not recognized before" is removed from that sentence.

My understanding of the "lack of mechanistic understanding" that the authors referred to is that it is more an issue of how to diagnose the reasons that the model cannot reproduce the observed effects of drought on ozone and aerosols. Drought affects pollutant concentrations through meteorological processes represented by variables such T, RH, and wind speed. There are many papers discussing these "mechanisms". Some are already referenced. But as I wrote in the initial review, the more relevant recent papers were not referenced. Zhang and Wang (2016) discussed the collinear problem in the correlations of ozone with T and RH (as was also seen in this paper) and ozone sensitivities to isoprene emissions. Ozone is much more sensitive to isoprene emissions in the fall than the summer. It is obviously relevant to the discussion of isoprene emissions in this paper.

The reviewer's point is well taken. We've added a new section (Section 3.2) to discuss extensively the meteorological factors responsible for the drought-pollutant relationship, such as temperature, RH, and wind speed. More relevant recent papers have been added as references, including Zhang and Wang (2016) and Zhang et al. (2017) mentioned by the reviewer. We acknowledge in this new section that there are well-established linkages between air quality and some meteorological parameters (e.g. temperature), thus the drought-pollution association may be partly explained by the effects of drought on these meteorological variables.

Zhang et al. (2017) showed that ozone high extremes are more likely to co-occur with high T and low RH. But they also showed that PM2.5 high extremes co-occur with high T and low wind speed but do not depend as much on RH in spring and fall. Drought events have high T, low RH, and low wind speed. Therefore, a drought index, which is more related to RH and T than wind speed, is not the most optimal variable to define the effects of meteorology on PM2.5 in seasons other than summer. The model errors that the authors referred to may be related to model biases in wind speed simulations under high T and low RH conditions (i.e. simulated drought).

In the new section 3.2, we added discussions of the differences between drought and meteorological factors (temperature, RH, and winds) and other meteorological events, including heat wave and stagnation, which are associated with high pollution levels and likely co-occur with drought. The first difference is that drought is not a daily-scale extreme or variable, such as temperature or RH. Drought arises only after a prolonged (> week) period of precipitation shortage that causes soil to dry up. Therefore, we chose the monthly scale to identify the drought-pollution association, differentiating it from day-to-day variability of meteorology. Second, drought is a complex extreme not based on individual meteorological parameters (e.g. temperature, humidity) or a simple combination of them. The prominent feature of drought is water deficit in both the atmosphere and the land component (e.g. soil and vegetation), resulting from the combination of precipitation shortage and increasing evapotranspirative water loss driven in part by high temperatures. As a result, the associated vegetation responses are likely to be more pronounced during drought than those associated with short-term meteorological extremes/events, which are relevant to our later discussion of isoprene changes.

We have added a clear statement to acknowledge that the co-occurrence of high temperature and low RH with drought is an important reason to explain the pollutant enhancements during drought, especially for surface ozone. However, it would not be feasible to separately quantify the effects of certain meteorological variables on the drought-pollution association, such as temperature, precipitation, and RH, because these variables are all factored in when defining drought. But wind speed is not an explicit factor in drought indices, thus we can evaluate if wind is a compounding meteorological factor for the drought-pollution association. The correlations (r) of monthly mean wind speeds with the SPEI (see new Figure S4 in supplementary material) are positive but small for the most part of the US ($r^2 < 0.2$). This suggests that wind speeds might not be an important meteorological factor responsible for the pollution enhancements during drought, except for localized areas where wind-blowing dust would be substantially higher during drought.

Based on Zhang et al. (2017), I suspect that ozone concentrations have a better correlation with a drought index than PM2.5 in spring and fall, even though the slope seems steeper for PM2.5 than ozone as a function of a drought index. This is obviously important in how the regression slopes can be used to infer the effects of drought. I already suggested in the initial review that the authors include a figure for the distributions of correlation coefficients of ozone and PM2.5 with a drought index (akin to Fig. 1). I make the same recommendation here.
The original manuscript showed the distributions of correlation coefficients in the supplementary material. We've now moved those figures to the main manuscript (new Figure 1). The correlation coefficients have similar spatial distributions as the correlation slopes for both ozone and $PM_{2.5}$.

It would take another paper to sort out all the details of why drought conditions lead to higher ozone and PM2.5 concentrations. That is not what I suggest that the authors do in this paper. But the relevant discussion suggested above should be included in the paper. Grouping data in summer with those in spring and fall is not a good choice (Zhang et al., 2017). Analyzing the data in summer and spring+fall separately will be much better. It may be a large amount of work, so I leave the choice to the authors. The authors may choose not to redo the analyses based on season. It is fine with this reviewer as long as the discussion of this seasonal issue is added.
The reviewer's point is well taken. We've added separate analysis of the ozone and $PM_{2.5}$ enhancement by season (spring, summer, and fall). See the new Figure 2 and related discussion added at the end of the revised Section 3.1. For ozone, all the regions see larger ozone enhancements in summer (Jun-Aug) and fall (Sep-Oct), while the spring (Mar-May) enhancement is the smallest. The seasonal differences of $PM_{2.5}$ enhancements are not statistically significant for most regions, nor are they coherent between regions, probably due to the complexity in $PM_{2.5}$ chemical constituents and sources. The seasonal comparison for a given region is based on the same sets of surface sites that experience droughts in all the seasons, thus the differences presented in the revised manuscript are not caused by sampling differences. The seasonal analysis supports the robustness of the drought-pollution association derived over the growing season as a whole.

References

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Zhang, Y., and Y. Wang, Climate driven ground-level ozone extreme in the fall over the Southeast United States, *Proc. Natl. Acad. Sci.*, 113, 10025–10030, doi: 10.1073/pnas.1602563113, 2011