Dear Dr. Alex (Co-editor),

I hereby submit final author responses to the referee comments and the revised manuscript on behalf of all the coauthors. We find that the comments of the three anonymous referees are well thought and very constructive. Below include a point-to-point response to the reviewer's comments in blue. The revised manuscript has take into account all the comments, including the following additional analysis and discussion:

(1) We added a new section (3.2) to discuss in depth the meteorological factors that may contribute to the drought-pollutant relationship (see detailed responses to referee 2 and 3);
(2) In that new section, we analyzed the co-occurrence of drought with stagnation and heat waves and quantified the impact of such co-occurrence to the drought-pollutant relationship. The results show more frequent stagnation and heat waves could explain up to 40% of the ozone and PM2.5 enhancements during drought, a significant but not majority factor (see detailed response to referee 3);
(3) We have separately analyzed the drought-pollutant relationships by season and by drought stage (see detailed response to referee 1 and 2);

I thank you and the three referees for the professional efforts in evaluating our manuscript and for the constructive comments to improve its quality. I look forward to hearing from you.

Best Regards,

Yuxuan (On behalf of all co-authors)

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 #1

This study addresses the effects of drought on air quality in the United States through statistical analysis of historical observations at surface monitoring sites and two drought indices, the Standardized Precipitation Evaporation Index (SPEI) and the

Palmer Drought Severity Index (PDSI). It also examines the ability of several current climate-chemistry models to simulate observed responses of ozone and fine particulate matter under drought conditions as identified by model-derived SPEIs. Future model projections of SPEI

and air quality are examined as well. The relationship of drought and air quality is a timely, highly relevant topic, appropriate for the readership of Atmospheric Chemistry and Physics.

Overall, the manuscript is generally well-written with only a few minor typographical or grammatical areas. There are several technical questions/comments that should be addressed prior to reconsideration for publication:

(1) Have many previous studies examined relationships between drought indices and observed air quality? Previous studies should be identified and to the extent possible discussed in the context of this work. See for example, Tian et al. doi:10.1002/ehs2.1203.

To our knowledge, few studies have examined the relationship between drought indices and observed air quality at a temporal and spatial scale similar to our study (i.e. 25 years, continental US). There are a few papers analyzing on one or two aspects of the drought impact on atmospheric compositions associated with dust and fire emissions (Prospero and Lamb, 2003;Westerling and Swetnam, 2003). Tian et al (2016) analyzed the combined effects of drought and ozone on crop productions in China, but they did not explicitly consider the drought effects on ozone. Our previous work (Wang et al., 2015) conducted a case study of surface $PM_{2.5}$ enhancements associated with the 2011 southern US drought. We have added discussions of all these previous studies in the introduction of the revised manuscript.

(2) Many drought indices exist now and the number will likely further evolve in the future. Are there indices that are particularly relevant for examining the relationship between drought and air quality, and if so why?

Air quality responds to changes not only of the atmosphere but also the land biosphere, thus the drought indices that are most relevant for air quality would be those that measure both meteorological (e.g., temperature and precipitation) and land biosphere conditions (e.g., soil moisture, evapotranspiration, vegetation, etc.) associated with drought. In addition, the temporal duration of drought is a matter of concern for air quality because air pollutants have different characteristic time scales with respect to transport and chemistry. This requires the relevant drought indices to be explicit of drought duration (e.g., month, year) in their calculation.

Take the Standardized Precipitation Evapotranspiration Index (SPEI) as an example, which is the primary drought index used in our study. The SPEI is based on water balance between precipitation and reference evapotranspiration, the latter dependent on atmospheric water demand related to temperature. Therefore it represents both meteorological conditions and water stress on land biosphere conditions during drought. In addition, the SPEI is multi-temporal and can specify drought duration of monthly, and multi-months. Our study used the 1-month SPEI and the correspondent monthly-mean air pollutant data (ozone and PM2.5) to derive the relationship between drought and air quality. By comparison, the Standard Precipitation Index (SPI) or the PDSI would not be a good drought index for air quality purpose because the SPI considers only meteorology (i.e. precipitation) while the PDSI does not specify drought duration.

The above points were implicit in the original manuscript where the SPEI is introduced (Section 2.1). We've now explicitly expressed them in the revised Section 2.1.

(3) Is there evidence in the historical data that the timing of the onset of drought influences air quality (e.g., late spring vs. early summer vs. late summer)? Is there evidence that prolonged drought more strongly influences air quality over time?

We added a new Figure 2 comparing the different effects of drought onset and prolonged drought on ozone and $PM_{2.5}$ enhancements. Both pollutants show larger enhancements during prolonged drought compared to drought onset across the four regions, except for $PM_{2.5}$ over the northeast. See the detailed discussion about Figure 2 added at the end of Section 3.1.

(4) More explanation as to how model-derived SPEIs were calculated (e.g. what method in the R package was used to determine PET?) and their performance relative to the global SPEI dataset and to each other would be beneficial. Model-derived

SPEIs are important to establishing predicted air quality during drought versus nondrought conditions and evaluating model deficiencies relative to observed responses in this work.

The model-derived SPEI were calculated with R package provided by the SPEI developer using model precipitation and temperature as inputs. The SPEI is derived as logistic-normalized distribution of water deficit, estimated as the difference between precipitation and reference evapotranspiration. Both Thornthwaite (Th) and Penman-Monteith (PM) method can be applied for estimation of the reference evapotranspiration. The Thornthwaite (Th) method only requires temperature data while the Penman-Monteith (PM) method requires additional inputs including RH, wind speed and radiation. Since ACCMIP model archives do not have all the variables required for the PM method, we used the Th method to calculate model SPEI. The global SPEI dataset use the PM method to estimate reference evapotranspiration. The correlation between SPEIs derived with PM and Th method is high (correlation r >0.9) (Beguera et al., 2014), thus the use of Th method may not have large impact on model SPEI calculation. We've clarified this point in the manuscript (Section 2.3).

With regard to the model ability of simulating drought, Figure S7 in the supplementary material presents the model-simulated drought frequencies during the historical period (1990-2014). The models can capture well 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. We've added discussion of the model SPEI in the manuscript (pg 9, line 33-39).

(5) It is acknowledged in the manuscript that the ACCMIP models vary widely in their predicted responses of air quality to drought. More explanation is needed regarding differences in the configuration and input data resources that could contribute to differences in their performance. A key outcome of this study should be to recommend specific paths forward for research that could lead to improvements in chemistry-climate model performance.

Agreed. Emissions, deposition, and chemistry are the most important aspects of model configurations affecting the drought-pollutants relationship. Anthropogenic emissions and biomass burning emissions were specified, but natural emissions were not, so the models treated natural emissions differently, which is a key factor in the different performance between models. For example, only the GISS-E2-R model simulates isoprene emissions as coupled with its meteorology (mostly temperature), thus allowing for isoprene emissions to increase with increasing temperatures. The other three models used prescribed BVOC emissions, thus representing different responses of those emissions to meteorology and climate change. All the ACCMIP models include dry and wet deposition of pollutants. While they all show large reductions of wet deposition during drought, the dry deposition is not sensitive to drought. With regard to aerosol chemistry, all the models overestimate the sulfate reduction, but at the same time underestimate the OA increase during drought. Both problems might be caused by the model misrepresentation of cloud sensitivity to changing drought severity, although the OA bias could also be caused by uncertainties of fire and BVOC emissions in the models. We've expanded the modeling discussion in Section 3.4 (last paragraph).

(6) Table 1, Fig. 2, Table S2 etc suggest that there are regional differences in contributions to drought effects to air quality, but the discussion is too limited in this regard. Are there opportunities to better understand model performance via examining regional responses?

This is a good point. We've expanded the discussion of regional differences in the revised Section 3.1 when presenting the regional-mean pollutants enhancements associated with drought (e.g. new Figure 2), as well as in the newly added Section 3.2 when presenting regional differences in meteorology during drought (e.g. new Figure 3 and Figure S4). A detailed region-to-region comparison is however outside the scope of the current manuscript and will be a future endeavor, as our main goal here is to provide observational evidence of the robustness and spatial prevalence of pollution enhancements during drought across the US.

Minor corrections:

First paragraph, introduction: Line 2: "matters" should be matter;

Line 4: missing "the" at the of the line; Line 10: missing noun after "recurring", Line 11: missing "the" before "atmosphere". Page 6, Line 2: "primarily resulted from" should be "primarily result from"

All are corrected.

References:

Beguera, S., Vicente-Serrano, S. M., Reig, F., and Latorre, B.: Standardized precipitation evapotranspiration index (SPEI) revisited: parameter fitting, evapotranspiration models, tools, datasets and drought monitoring, *International Journal of Climatology*, 34, 3001-3023, 2014.

Prospero, J. M., and Lamb, P. J.: African droughts and dust transport to the Caribbean: Climate change implications, *Science*, 302, 1024-1027, 2003.

Tian, H., Ren, W., Tao, B., Sun, G., Chappelka, A., Wang, X., Pan, S., Yang, J., Liu, J., and S Felzer, B.: Climate extremes and ozone pollution: a growing threat to China's food security, *Ecosystem Health and Sustainability,* 2, 2016.

Wang, Y., Xie, Y., Cai, L., Dong, W., Zhang, Q., and Zhang, L.: Impact of the 2011 southern US drought on ground-level fine aerosol concentration in summertime, *Journal of the Atmospheric Sciences*, 72, 1075-1093, 2015.

Westerling, A. L., and Swetnam, T. W.: Interannual to decadal drought and wildfire in the western United States, *EOS, Transactions American Geophysical Union*, 84, 545-555, 2003.

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

Zhang, H., Y. Wang, T.-W. Park, and Y. Deng, Quantifying the relationship between extreme air pollution events and extreme weather events, *Atmos. Res.*, 188, 64-79, doi:10.1016/j.atmosres.2016.11.010, 2017.

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, 20

Reviewer #3

This study examines the correlations between drought and air pollutants (ozone and

PM2.5) in the US. The authors use the linear regression slope derived from drought indices and ozone/PM observations to infer the effects of drought and argue that most chemistry-climate models are not able to reproduce the observed relationships. The authors further apply the observed relationships to climate model projected drought occurrences and attempt to estimate the effects of increasing drought on ozone and PM by 2100 compared to the 2000s.

The manuscript is well structured and readable. However, there is a major flaw in the applied method to quantify the impact of drought. The correlations between drought and ozone reported in this study may reflect the common underlying correlation with air stagnation and temperature rather than a causal relationship of drought on ozone. An inspection of the model differences in their Figure 5 supports this statement. None of these models include the effects of soil moisture deficits on BVOC emissions and the reduced efficiency of ozone dry deposition sink to vegetation. Nevertheless, the GISS model with interactive isoprene emissions simulates the SPEI/ozone slope comparable to the observed values over the Southeast. The greater slope simulated in GISS as compared to other models reflects the inclusion of interactive isoprene emissions, which allows the model to simulate ozone enhancements resulting from stronger isoprene emissions during heat waves (see Schnell et al., 2016). Reduced BVOC emissions under drought stress will actually lead to less ozone.

We thank the reviewer for making this important point, which we have careful analyzed and extensively discussed in the revised manuscript. We've added a new session (Section 3.2) and several new figures (Figure 3, Figure S4-S6) to discuss the meteorological factors behind the SPEI-pollutant relationship and the possible compounding effects of stagnation and high temperatures.

In the new Section 3.2, we first acknowledge 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. We then discussed the differences between drought and meteorological parameters (temperature, RH, and winds) and meteorological events, including heat wave and stagnation. 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. The change in isoprene during drought found in our study is consistent with Schnell et al. (2016) in that isoprene tends to increase in most drought conditions (Figure 4); only under extreme drought (SPEI <-2) isoprene is shown to decrease in limited observations.

We then examined the relationships of monthly occurrences of stagnation and heat waves with the SPEI at each 0.5^{o}x0.5^{o} grid over the study period; the results are shown in the new Figure S4 in the supplementary material. There are positive correlations between drought-stagnation and drought-heatwave across the US, indicating both events do occur more frequently during drought

months. But the squares of these correlations are all below 0.4, with a typical value of 0.1-0.2 for the most parts of the US. This suggests that on the monthly scale stagnation and heat waves would typically be able to explain 10%~20% variability in the SPEI, a non-trivial but small fraction. As shown in the lower panel of Figure S4, stagnation and heat waves have an average 7% and 5% increase in their frequencies during drought months compared to normal months, although the extent of such increases varies greatly by region. The maximal increase of stagnation frequency during drought is about 15% in the west, southern Great Plains and southwest, where stagnation tends to occur frequently even during normal conditions. The largest increase of heat waves during drought is about 20% in the southern Great Plains.

Finally, to quantify the compounding effects of stagnation and heat waves on the drought-pollution association, we re-evaluated the SPEI-pollutant relationships by applying weights to each pair of SPEI and pollutant anomalies (ozone and $PM_{2.5}$). The weights are given as the percentages of days in each month (regardless of drought or non-drought) that are neither stagnation nor heat wave, assuming the two events are mutually exclusive which would give an upper bound for the weights. Since the weights are between 0 and 1, the weighting process effectively scales down the magnitude of pollution anomalies in each month. The weighted correlation and enhancements are shown in the new Figure 3. The weighting does not change the sign or statistical significance of the SPEI-pollutant correlations at all the sites, indicating the covariance of drought with stagnation and heat waves might not be the dominant factor causing the drought-pollutant correlations. The weighting however reduces the magnitude of ozone and $PM_{2.5}$ enhancements associated with drought by an average of 40% when both events are combined. This indicates that more frequent stagnation and heat waves could explain up to 40% of the ozone and $PM_{2.5}$ enhancements during drought, a significant but not majority fraction.

While severe drought can potentially lead to elevated surface ozone by reducing the ozone dry deposition sink to vegetation (see a review by Fowler et al., 2009), this impact has to be demonstrated using a more sophisticated statistical approach (e.g.,

multi-variate regression) or chemistry-climate model sensitivity experiments to isolate the role of air stagnation and temperature. For example, Lin et al. (2017) showed that reducing ozone Vd by 35% in GFDL-AM3 during the severe North American drought of 1988 simulates 10 ppbv greater ozone enhancements than a BASE simulation with constant Vd, although the BASE simulation still captures observed ozone enhancements during the other warm summers driven by processes other than drought (see their Section 6 and Figs.18 and 19).

We agree with the reviewer that this manuscript is not an attribution analysis (e.g. using chemistry-climate model sensitivity experiments), and we have explicitly stated so in the manuscript: "… attribution of the underlying causes would require chemistry-climate model sensitivity experiments, which is outside the scope of the present study" (pg 10, line 33-36). We've added Lin et al. (2017) as reference when discussing the dry deposition effect.

Without a more careful attribution analysis to separate the influence of stagnation and temperature, you cannot use the terms like “drought-induced”, “causes of ozone and PM enhancements by drought” or “effects of droughts”. All such terms in the present manuscript will need to be removed or rephrased.

Our new analysis (Section 3.2) showed that the drought-pollution relationship is still significant after discounting the influence of stagnation and temperature on air pollution (see the response to the first comment), which supports our argument that droughts affect air quality. However, we agree with the reviewer that our manuscript is not an attribution analysis by design and thus some of the terms need to be modified. We’ve modified all the attribution terms throughout the manuscript to non-attribution ones such as “pollutant enhancement associated with droughts” or “drought-pollutant relationship” or “drought-pollutant association”.

In summary, the analysis presented in the current manuscript shows a correlation between drought indices and air pollutants but not the causal effects of drought on air quality. The derived slope may serve as a useful diagnostic to evaluate the models, as the authors show, but it cannot be used to quantify the impact of drought on air quality.

We agree. As stated above, we’ve changed attribution statements to non-attribution ones throughout the manuscript.

Relevant references:

Fowler D, Pilegaard K, Sutton M, Ambus P, Raivonen M, Duyzer J*, et al.* Atmospheric composition change: ecosystems–atmosphere interactions. *Atmospheric Environment* 2009, **43**(33)**:** 5193-5267.

Lin, M., Horowitz, L. W., Payton, R., Fiore, A. M., and Tonnesen, G.: US surface ozone trends and extremes from 1980 to 2014: quantifying the roles of rising Asian emissions, domestic controls, wildfires, and climate, *Atmos. Chem. Phys*., 17, 2943-2970, https://doi.org/10.5194/acp-17-2943-2017, 2017.

Schnell JL, Prather MJ, Josse B, Naik V, Horowitz LW, Zeng G*, et al.* Effect of climate change on surface ozone over North America, Europe, and East Asia. *Geophysical Research Letters*, 43(7)**:** 3509-3518, 2016

Kavassalis, S.C. and Murphy, J.G.: Understanding ozone-meteorology correlations: A role for dry deposition. *Geophysical Research Letters*, 44(6), pp.2922-2931, 2017

Adverse Effects of Increasing Drought on Air Quality via Natural Processes

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Abstract. Drought is a recurring extreme of the climate system with well-documented impacts on agriculture and water resources. The strong perturbation of drought to the land biosphere and atmospheric water cycle will affect atmospheric composition, the nature and extent of which are not well understood. Here we present observational evidence that ~~surface ozone and $PM_{2.5}$ in the US are~~US air quality is significantly correlated with drought severity. Severe droughts during the period of 1990-2014 were found associated~~,~~ with growth-season (Mar-Oct) mean enhancements in surface ozone and $PM_{2.5}$ of 3.5 ppbv (8%) and 1.6 μg m^{-3} (17%)~~-~~, respectively~~increases respectively under severe drought~~. ~~These~~ The pollutant enhancements associated with droughts do not appear to be affected by ~~show little sensitivity to~~ the decreasing trend of US anthropogenic emissions, indicating natural processes as the primary cause. Elevated ozone and $PM_{2.5}$ are attributed to the combined effects of drought on deposition, natural emissions (wildfires, biogenic VOCs and dust), ~~deposition,~~ and chemistry. Most climate-chemistry models are not able to reproduce the observed ~~responses~~ correlations of ozone and $PM_{2.5}$ to drought severity~~, suggesting a lack of mechanistic understanding of drought effects on atmospheric composition~~. The model deficiencies are partly attributed to the lack of drought-induced changes in land-atmosphere exchanges of reactive gases and particles and misrepresentation of cloud changes under drought conditions. By applying the observed relationships between drought and air pollutants to climate model projected drought occurrences, we estimate an increase of 1-6% for ground-level O_3 and 1-16% for $PM_{2.5}$ in the US by 2100 compared to the 2000s due to increasing drought alone. Drought thus poses ~~another~~ an important aspect of climate change penalty on air quality, and a ~~not recognized before. Improvements in the models are imperative to facilitate~~ better prediction of ~~air quality challenges due to changing hydroclimate and atmospheric composition feedback to climate~~such effects would require improvements in model processes.

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1. Introduction

Air pollution is a major global health risk (Forouzanfar et al., 2015). Chronic and acute exposure to enhanced ozone (O_3) and fine particulate ~~matters~~ matter with diameters less than 2.5 μm ($PM_{2.5}$) has been associated with many adverse health impacts and premature mortality (Lelieveld et al., 2015). Ambient O_3 and $PM_{2.5}$ concentrations are strongly regulated not only by the atmosphere but also by land-atmosphere interactions through emission and deposition processes. To date, the variation of air quality with climate change has not been fully revealed as most analysis in the past were conducted with respect to atmospheric parameters or events only, such as temperature (Steiner et al., 2010), precipitation (Dawson et al., 2007; Allen et al., 2015), and short-term (in the order of days) meteorological anomalies (e.g., heat/cold waves, air stagnation, and temperature inversion) (Filleul et al., 2006; Qu et al., 2015; Hou and Wu, 2016). The impact of changing hydroclimate on air pollution is largely unexplored and highly uncertain, particularly with respects to droughts, a type of complex extremes ~~recurring in many parts of the world that could affect atmosphere, land, and their interactions from weeks to seasons and even years~~on the time scale of weeks to months or longer which affect not only the atmosphere but also its interactions with the land biosphere.

Drought is characterized by a prolonged period of precipitation shortage and soil moisture deficit in combination with high temperatures (Trenberth et al., 2014). Drought impacts on agriculture and water resources have been extensively documented ~~Besides significant impacts on agriculture and water resources~~ (Rosenzweig et al., 2001; Arnell, 2004). With regard to air pollution, drought ~~conditions affect precipitation scavenging, chemical production/loss, and hence atmospheric lifetime of atmospheric constituents~~can reduce wet scavenging of pollutants, affect their chemical production/loss, and change their atmospheric lifetime. Drought also influences ~~(Wang et al., 2015). Through changing~~ the health and conditions of soil and vegetation cover across the landscape, ~~drought can perturb~~thus perturbing upward transmission of dusts (Prospero and Lamb, 2003) and reactive gases (e.g., biogenic volatile organic compounds or BVOCs and NO_x) (Fuentes et al., 2000; Guenther et al., 2012; Guenther, 2015; Davidson et al., 2008) from the surface into the atmosphere as well as downward dry deposition of gases and aerosols (Huang et al., 2016). Complications such as increasing wildfires and changing human activities (Westerling et al., 2003; Scanlon et al., 2013) further compound the effects of drought on atmospheric composition. Previous studies illustrated one or two aspects of the complex effects of drought on atmospheric composition. For example, Westerling et al. (2003b) and Prospero and Lamb (2003) explored the potential risks of increasing wildfire and dust emissions under drought conditions. Tian et al (2016) analysed the combined effects of increasing drought and increasing ozone levels on crop production in China, although in their study the ozone change was not linked to drought change. Our prior analysis (Wang et al., 2015) showed a 26% enhancement of surface $PM_{2.5}$ concentrations in the southern US during the severe summer drought conditions in 2011, and suggested wildfires and cloud processes as key factors responsible for the change of $PM_{2.5}$ during drought. A comprehensive assessment of air pollution changes during different drought periods has yet to be conducted to verify the findings from case studies and to reveal important processes responsible for the associated changes. In addition, c~~C~~limate change has the potential to increase the frequency and magnitude of droughts in many parts of the world (Dai, 2012; Cook et al., 2015), further underscoring the importance of understanding the full extent of drought impacts.

In this study we first quantify the impact of historical droughts on air quality, an area largely overlooked in prior investigations of drought impacts, and discuss the possible causes of those impacts. The regional focus is the continental US where observational records of atmospheric composition are most abundant. The study period is 1990-2014 and the growing season (March-October) when drought has most deleterious impacts on the land and biosphere. We then assess the performance of current

chemistry-climate models in capturing the response of surface air pollutants to drought. Future changes in air quality related to increasing drought are estimated.

2. Data and Method

2.1 Drought index

~~Among~~ There are many types of drought ~~indicators~~indices (Heim and Richard, 2002). The drought indices most relevant for air quality would be those capable of representing both meteorological (e.g., temperature and precipitation) and land biosphere conditions (e.g., soil moisture, evapotranspiration, vegetation, etc.) associated with drought, because air pollution levels are dependent not only on meteorology but also the land-atmosphere interaction. In addition, air pollutants have various characteristic time scales, thus the relevant drought indices should be able to specify the duration of droughts.~~,~~ Here we chose the standardized precipitation evapotranspiration index (Vicente-Serrano et al., 2010) (SPEI) to examine the relationship between drought and air quality. ~~because~~ The SPEI~~it~~ is ~~multiscalar~~ multi-temporal in representing the ~~drought~~ duration of drought and its formulation is based on water balance approach which explicitly considers the impact of temperature variations on evaporation. To identify the full extent of drought impacts and differentiate drought from normal variability in the hydrological cycle, we used the 1-month SPEI to select droughts lasting more than one month. ~~explicitly considers the impact of temperature variability on water balance.~~ The gridded SPEI datasets are obtained from the global SPEI database (http://sac.csic.es/spei/) with a spatial resolution of 0.5 ^{o} x 0.5^{o}. While ~~The study period is 1990-2014 and the regional focus is the continental US where observational records of atmospheric composition are most abundant. We focus on the growth season (March-October) during which drought has most deleterious impacts on the land and biosphere. To identify the full extent of drought impacts and differentiate drought from normal variability in the hydrological cycle, we used the 1-month SPEI to select droughts lasting more than one month.~~ n~~N~~egative SPEI typically indicates drought~~.~~~~,~~ a~~A~~ more strict criteria of SPEI < -1.3 (the lowest 10th percentile of SPEI) was used here to distinguish drought conditions from non-drought conditions (SPEI between -0.5 to 0.5).

To test the robustness of the drought-pollution relationship derived from SPEI, we used the Palmer Drought Severity Index (PDSI) to evaluate this relationship. The PDSI is the most widely used index of meteorological drought in the US and best represents long-term drought (~12 months) (Heim et al., 2002). Among all forms of PDSI, sc_PDSI_pm is the most updated version with self-calibration and improved formulation of calculating potential evapotranspiration (Dai, 2011). The sc_PDSI_pm dataset (assessed from http://www.cgd.ucar.edu/cas/catalog/climind/pdsi.html) is monthly with a spatial resolution of 2.5 ^{o} x 2.5^{o} ~~(Assessed from http://www.cgd.ucar.edu/cas/catalog/climind/pdsi.html)~~. Drought conditions were identified when sc_PDSI_pm < -3.

Figure 1a shows the percent occurrence of drought months (SPEI < -1.3) over the continental US during the study period. The Western US, Great Plains, Southeast US and southern part of the Northeast US clearly stand out as the most drought prone regions, with extreme droughts occurring 10%-25% of time, ranging between 20 and 40 months during the past 25 years (Figure 1b). Recent examples of infamous droughts are the 2011 Texas drought (Nielsen-Gammon, 2012), the 2012 Great Plains drought (Hoerling et al., 2014), and the 2014-2015 California drought (Griffin and Anchukaitis, 2014). The PDSI-derived drought occurrence frequency (sc_PDSI_pm<-3; Figure S1) shows a similar pattern. However, the areas with more than 10% drought occurrence based on sc_PDSI_pm are much smaller than those based on SPEI (Figure S1a). This is partly because the two indices represent drought at different time scales (i.e. one month for SPEI versus 12 months for sc_PDSI_pm).

2.2 Air pollution and meteorological data

Surface concentrations of $PM_{2.5}$ and maximum daily 8 hour running average (MDA8) ozone over the same period were derived from daily observations collected over more than 2000 surface sites from the US Environmental Protection Agency Air Quality System (EPA-AQS) (http://aqsdr1.epa.gov/aqsweb/aqstmp/airdata/download_files.html), Clean Air Status and Trends Network (CASNET; https://www.epa.gov/castnet), and the Interagency Monitoring of Protected Visual Environments (IMPROVE) (Malm et al., 1994, http://views.cira.colostate.edu/) networks. Those daily observations were averaged to monthly means for analysis. The site-specific SPEI is the SPEI at the grid containing each site. Speciated $PM_{2.5}$ data was obtained from the Speciation Trend Networks (STN), which is a subset of the EPA AQS with about 180 sites. Sulfate wet depositions were collected from the National Atmospheric Deposition Program (NADP; http://nadp.isws.illinois.edu/). Isoprene concentrations were obtained from the Photochemical Assessment Monitoring Stations (PAMS) network (https://www3.epa.gov/ttnamti1/pamsdata.html). Surface data at each site was deseasonalized and detrended by removing the 7-year moving averages from the raw data time series for each month to derive the anomalies (c.f. Figure S2 for an example of data processing). The relationship between SPEI and air pollution anomalies was calculated by linear regression, and the *p* values are obtained from two-tailed *F*-test. Regional analysis focuses on four geographical divisions of the continental US (Figure 1a): the Western US [128°W-106°W, 30°N-50°N], the Great Plains [106°W-96°W, 25°N-50°N], the Southeast US [96°W-75°W, 25°N-38°N] and the Northeast US [96°W-63°W, 38°N-50°N].

Fire emissions were obtained from the Global Fire Emission Database (GFED) at a 0.25^o x 0.25^o resolution (Giglio et al., 2013; Randerson et al., 2012; Van der Werf et al., 2010; Akagi et al., 2011; http://www.falw.vu/~gwerf/GFED/GFED4/). The spatial impacts of fire smokes can range from a few kilometers to thousands of kilometers depending on the burning area/intensity, injection height, and transport conditions. Fire emissions from 9 grid points (~40 km) around each surface site were sampled to represent the immediate and transported impacts of fires. Temperature and~~,~~ precipitation~~, incoming shortwave radiation, and cloud fraction~~ were obtained from the ~~Modern Era Retrospective Analysis for Research and Applications (MERRA) reanalysis (Rienecker et al., 2011) (http://giovanni.gsfc.nasa.gov/giovanni/)~~Climatic Research Unit (CRU, v3.22), which were also used as input data for global SPEI dataset. Monthly mean cloud fractions ~~below 680 hPa~~ from satellite observations were obtained from the Clouds and the Earth's Radiant Energy System (CERES) ISCCP-D1~~2~~like products at a spatial resolution of 1^o x 1^o for the period of 2000 to 2014 (Minnis et al., 2011, https://ceres-tool.larc.nasa.gov/ord-tool/jsp/ISCCP-D1Selection.jsp~~https://eosweb.larc.nasa.gov/project/ceres/isccp-d2like-merged-ed3a-table~~). The site-specific meteorological parameters were retrieved from the grid that contains a surface site.

2.3 Models

We evaluated the SPEI-pollution relationships simulated by four models from the Atmospheric Chemistry and Climate Model Intercomparison Project (ACCMIP) (Lamarque et al., 2013) that have archived ozone and $PM_{2.5}$ concentrations: GISS-E2-R, GFDL-AM3, NCAR-CAM3.5, and MIROC-CHEM (Downloaded from http://browse.ceda.ac.uk/). The ACCMIP~~-~~ experiments were forced with observed greenhouse gases concentrations from historical runs. The four models used the same anthropogenic and biomass burning emissions of ozone and aerosol precursors (Lamarque et al., 2010). While anthropogenic emissions were yearly specific, biomass burning emissions were present at the decadal mean without inter-annual variations within a specific decade. Natural emissions were not specified, so the models treated natural emissions differently with different responses to drought. For example, isoprene, the most abundant BVOC, is an important precursor of tropospheric ozone and

secondary organic aerosols. Only the GISS-E2-R model simulates isoprene emissions as coupled with its meteorology (mostly temperature), thus allowing for isoprene emissions to increase with increasing temperatures. The other three models used prescribed BVOC emissions, thus representing different responses of those emissions to meteorology and climate change. ~~The treatment of natural emissions varies between models, depending on meteorological and surface conditions. The GISS-E2-R model simulates isoprene emissions as coupled with its meteorology, while the GFDL-AM3, NCAR-CAM3.5 and MIROC-CHEM model used prescribed emissions.~~ ACCMIP focuses on time-sliced experiments, thus each model covers different time periods. Model ozone and $PM_{2.5}$ were deseasonalized and detrended for each time slice experiment in order to remove the effect of changes in anthropogenic emissions.

The model SPEI was calculated using the R package provided by the SPEI developers (https://cran.r-project.org/web/packages/SPEI/index.html), with simulated precipitation and temperature from each model as inputs. Model temperature was used to estimate reference evapotranspiration using the simplified Thornthwaite (Th) method. The model SPEI was then derived based on logistic-normalized distribution of water deficit, which is the difference between the reference evapotranspiration and model precipitation. Although more accurate estimates of evapotranspiration can be derived using the more complicated Penman-Monteith (PM) method, as used in the historical SPEI database, it requires additional input data not available from the ACCMIP archive. The Th-derived SPEI is shown to have tight correlations with the PM-derived SPEI ($r > 0.9$) (Beguer á et al., 2014). ~~based on simulated precipitation and temperature from each model.~~ The relationship of SPEI with air pollution anomalies was derived over all the time periods with available model outputs. To evaluate cloud properties in the model, we used the random overlap approach (Stephens et al., 2004) to calculate the total cloud fraction (CF) (1000 hPa to 10 hPa) and boundary layer CF (1000 hPa to 800 hPa), which can be the relevant cloud property for tropospheric ozone photochemical formation and cloud processing of aerosols, respectively. ~~To evaluate low-altitude cloud fractions in the model, which is most relevant to atmospheric chemistry, we calculated the cloud fraction below 680 hPa using the random overlap (Stephens et al., 2004).~~ Further details on the model experiments and data processing are listed in Supplementary Table S1.

3. Retrospective Analysis

3.1 Association between drought and air pollution

~~**3.1 Effects of drought on air pollutants**~~

We first derived the general association of surface ozone and $PM_{2.5}$ with the SPEI at the surface sites. ~~(Figure 1)~~. ~~Concentrations at each surface site were deseasonalized and detrended (as described in Section 2.2)~~ ~~T~~to remove the effects of seasonality and long-term changes in anthropogenic emissions, pollutant concentrations at each surface site were deseasonalized and detrended (as described in Section 2.2), and the resulting anomalies were used for analysis. Ozone and $PM_{2.5}$ anomalies show spatially prevalent negative correlations with the SPEI (Figure 1), with statistically significant correlations ($p < 0.05$) at 75% - 88% of the sites. Since negative SPEI indicates drought, the negative correlations indicate higher pollution levels during drought.~~statistically significant and spatially prevalent negative correlations with SPEI (Figure S3) at 75% - 88% of the sites.~~ The slopes from linear regression span a range of -4.75~-0.42 for the ozone-SPEI relationship and -2.2~-0.15 for that of $PM_{2.5}$-SPEI, with the mean of -2.21 and -0.83, respectively. The mean regression slope could be interpreted to indicate that an increase in drought severity by one standard deviation of the SPEI is associated with~~suggest larger sensitivities to drought for both ozone (-2.63) and $PM_{2.5}$ (-1.0) in the east (Figure 1c, e). These slopes~~

~~indicate that decreasing SPEI by one standard deviation will lead to~~ an average increase of 2.21 (±0.85) ppbv for ozone and 0.83 (±0.37) μg m^{-3} for $PM_{2.5}$ in the US. Both slopes are higher in the east, suggesting larger sensitivities to drought for both ozone (-2.63) and $PM_{2.5}$ (-1.0) (Figure 1d and 1g). Consistently with the SPEI, the PDSI shows statistically significant negative correlations with ozone and $PM_{2.5}$ anomalies at the majority of the sites (Figure S1).~~The slope from linear correlation between sc_PDSI_pm and ozone and $PM_{2.5}$ are shown in Figure S1.~~ The correlations with the PDSI are nevertheless weaker, because the SPEI is more suitable to present drought at the monthly scale than PDSI.~~There are 52-74% sites showing negative correlations, a lower percentage compared to the correlations based on SPEI because SPEI has more accurate representation of drought at the monthly scale than sc_PDSI_pm.~~

To further distinguish the drought effects, we aggregated pollutant anomalies from the sites with greater than 10% occurrence of drought onto three dryness levels: drought (SPEI < -1.3), normal (SPEI between -0.5 and 0.5), and wet (SPEI >-1.3). The composite comparison of ozone and $PM_{2.5}$ between those dryness levels is shown in Figure 1e and 1h, respectively. Significantly higher levels of both pollutants are found associated with drought across all the regions. ~~This composite comparison reveals significantly higher levels of both pollutants during drought as compared to normal and wet conditions, which is evident consistently across all the regions (Fig. 1d, f). Average enhancement in the US is 3.5 (6.6) ppbv for ozone and 1.6 (2.5) μg m^{-3} for $PM_{2.5}$ under drought as compared to normal (wet) conditions.~~ The eastern sites show a larger ~~increase~~ enhancement of ozone and $PM_{2.5}$ during drought, consistent with the SPEI-pollutant regression slopes being highest in this region. The response of air pollution to drought can be quantified as the difference of pollutants (ozone and $PM_{2.5}$) anomalies during drought relative to their levels during normal conditions. This difference is referred to as enhancement because it is predominantly positive. The average of such enhancements in the US is 3.5 ppbv for ozone and 1.6 μg m^{-3} for $PM_{2.5}$. Despite regional differences in absolute pollution levels, the relative pollution enhancement during drought is similar across regions at about 8% for ozone and 17% for $PM_{2.5}$ (Figure S3). The enhancements reported hereafter are all evaluated relative to normal conditions unless noted otherwise; if relative to wet conditions, the magnitudes of the enhancements are typically about a factor of two higher.

~~ozone at 3.9 (7.3) ppbv and $PM_{2.5}$ at 2.0 (3.1) μg m^{-3} under drought as compared to normal (wet) conditions, consistent with the spatial distribution of the regression slopes. The relative change is similar across regions at about 8% (16%) for ozone and 17% (31%) for $PM_{2.5}$ (Figure S4).~~

The composite comparison based on the PDSI is displayed in Figure S1. Since drought frequency represented by the PDSI is comparatively lower, ~~The distribution of ozone and $PM_{2.5}$ anomalies at different dryness levels based on sc_PDSI_pm are displayed in Figure S1 (d and f). Since drought frequency represented by sc_PDSI_pm is comparatively lower,~~ we chose sites with more than 5% drought occurrence ~~based on sc_PDSI_pm~~ and 5 years of available surface observations to reduce the spatial sampling bias. The average enhancement associated with drought derived from the PDSI is 2.3 ppb for O_3 and 1.2 μg m^{-3} for $PM_{2.5}$ in the US. The relative enhancement is similar across different regions at about 5% for ozone and 14% for $PM_{2.5}$, smaller but consistent with the results based on the SPEI. Such consistency indicates that the association between air pollution and drought would not depend on one's choice of drought indicator. However, the SPEI is a more suitable index than the PDSI to identify the drought-pollution association at the monthly time scale, and the following analysis is all based on the SPEI.

To test the robustness of the drought-pollution association and explore the temporal characteristics of such association, we performed the composite comparison of air pollutants between drought and normal conditions separately by season (spring, summer, fall) and by drought stage (onset vs. prolonged). Drought onset is defined as the first month when a drought occurs at a given location; if a drought lasts only one month, that month is also labeled as onset. A prolonged drought is one when the proceeding

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month is also a drought. Figure 2 compares the variations of regional ozone and $PM_{2.5}$ enhancements during drought derived from different temporal sampling approaches. The growing season (Mar-Oct) mean enhancement of ozone is close to 3 ppbv in the west and Great Plains, increasing to 3.9 ppbv in the southeast and northeast. The same spatial gradient is found in the growing season mean enhancement of $PM_{2.5}$, which increases from a mean of 0.9 μg m^{-3} in the west and Great Plains to 2 μg m^{-3} in the southeast and northeast. Seasonally, all the regions see larger ozone enhancements in summer (Jun-Aug) and fall (Sep-Oct), with the spring (Mar-May) enhancement being the smallest. The Southeast and the Great Plains have the largest seasonal difference in the response of ozone to drought. Relative to the growing season mean, the ozone enhancements in those region are about 38% higher in summer/fall and 50% lower in spring. 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 (to be discussed in Section 3.3). Only the northeast shows a significantly larger $PM_{2.5}$ enhancement in summer and significantly smaller enhancement in spring, about 42% higher and 27% lower than the growing-season mean, respectively. The seasonal comparison for a given region is based on the same sets of surface sites that experienced droughts in all the seasons, thus the differences presented above are not caused by sampling differences.

With respect to drought duration (Figure 2), surface observations in all the regions reveal significantly larger enhancements of both ozone and $PM_{2.5}$ during prolonged drought months than the onset months, with the only exception of $PM_{2.5}$ in the northeast which shows a significantly higher enhancement during drought onset. The largest sensitivity to drought duration is found in the southeast, where both ozone and $PM_{2.5}$ enhancements are higher by up to 50% during prolonged droughts than the onset. Again, the differences shown for a given region are not caused by sampling differences, as the comparison is based on the same sets of surface sites.

3.2 Meteorological factors for the drought-pollution association

Before proceeding with a discussion of the causes of the drought-pollution association, it is useful to present the difference between drought and some meteorological conditions/extremes likely to co-occur with drought that are also associated with higher pollution levels. For example, high ozone is more likely to occur with high temperature and low RH (Hou et al., 2016; Zhang and Wang, 2016; Zhang et al., 2017), and during heat waves (Filleul et al, 2006), conditions often co-occurring with drought. High $PM_{2.5}$ events in the US are found to co-occur with high temperature and low wind speed, but not consistently dependent on RH (Tai et al., 2010; Zhang et al., 2017). Stagnation days typically result in high ozone and $PM_{2.5}$ levels at the surface (Tai et al., 2010; Schnell and Prather, 2016). Compared with those types of weather phenomena and extremes defined on the daily basis, drought has on a longer time scale of at least one week and often monthly. For example, one would not call it a drought if no rain for a few days. 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.

Furthermore, 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. During the historical drought periods analyzed here, the surface sites were affected with precipitation decreases of up to 50% regionally and temperature increases of up to 2 ^{o}C, as compared to normal conditions (Table S2). Large changes in other meteorological variables are also associated with drought conditions, such as a 10% decrease in RH, 39% decrease in cloud fraction, and an increase in incoming solar radiation by 12.4 W/m^2. Because the time scale of drought is monthly,

these meteorological changes are persistent changes on the monthly scale, as opposed to day-to-day variability. As a result, the associated vegetation responses are likely to be more pronounced during drought than those associated with short-term meteorological extremes/events, with important implications for the land-atmosphere exchanges of reactive gases and aerosols.

As discussed above, 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. For example, the co-occurrence of drought with high temperature and low RH 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 other meteorological variables might be ruled out as compounding the drought-pollution association. For example, wind speed is a key factor influencing air quality, but not an explicit factor in drought indices. The correlations (r) of monthly mean wind speeds from ERA-reanalysis with the SPEI (Figure S4) are positive but small for the most part of the US ($r^2 < 0.2$), except for the northwest corner and surroundings of the Great Lakes with r^2 of 0.3~0.4. 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 become substantially higher during drought.

In addition, drought months may consist of a larger number of meteorological extremes conducive for high pollutant levels, such as stagnation and heat waves. To understand the pattern and extent of such co-occurrence, we examined the relationships of monthly occurrences of stagnation and heat waves with the SPEI at each 0.5^{o}x0.5^{o} grid over the study period (Figure S4). The frequency of stagnation was derived from the NOAA Air Stagnation Index (ASI, https://www.ncdc.noaa.gov/societal-impacts/air-stagnation/), in which stagnation is defined when surface wind speed smaller than 3.2 m/s, 500 mb wind smaller than 13 m/s and no precipitation, following Wang and Angell (1999). Heat waves were defined as two consecutive days with daily mean temperature greater than 90^{th} percentile of the warm-season (May to Sep) daily mean temperature during 1990-2014, following the method by Anderson (2011), using ERA Interim reanalysis (Dee et al., 2011) as inputs. Temporally, both stagnation and heat waves show negative correlations with the SPEI across the US (upper panel of Figure S4), an indication of more days of these meteorological extremes during drought months, but the squares of these correlations are all below 0.4, with a typical value of 0.1-0.2 for the most parts of the US. This suggests that on the monthly scale stagnation and heat waves would typically be able to explain 10%~20% variability in the SPEI, a non-trivial but small fraction. The exceptions are found in isolated locations in the west and southeast where stagnation could explain up to 40% of the SPEI variability, and the southern Great Plains with up to 30% of the SPEI variability explained by heat waves. Stagnation has an overall higher correlation with the SPEI than heat waves, partly because stagnation days by definition exclude precipitation. The lower panel of Figure S4 shows that stagnation and heat waves have an average 7% and 5% increase in their frequencies during drought months compared to normal months, although the extent of such increases varies greatly by region. The maximal increase of stagnation frequency during drought is about 15% in the west, southern Great Plains and southwest, where stagnation tends to occur frequently even during normal conditions. The largest increase of heat waves during drought is about 20% in the southern Great Plains.

To quantify the compounding effects of stagnation and heat waves on the drought-pollution association, we reevaluated the SPEI-pollutant relationships by applying weights to each pair of SPEI and pollutant anomalies (ozone and $PM_{2.5}$). The weights are given as the percentages of days in each month (regardless of drought or non-drought) that are neither stagnation nor heat wave, assuming the two events are mutually exclusive which would give an upper bound for the weights. For example, a month

with none of the two events is given 100% weight when calculating the SPEI-pollutant correlation and pollutant enhancement, while a month with 15 days of those events has a weight of 50%. The weighted enhancement is calculated as the difference in weighted-mean anomalies between drought and normal months. Since the weights are between 0 and 1, the weighting process effectively scales down the magnitude of pollution anomalies in each month, assuming the effects of stagnation and heat waves are linear to their occurrences. Figure 3 compares the original (un-weighted) and weighted correlations, regression slopes, and pollution enhancements. The differences in correlation coefficient (r) are mostly smaller than 0.05 in terms of absolute values. The exception is for ozone in the west where the absolute value of the weighted r is increased by 0.1~0.2, revealing a stronger correlation between SPEI and ozone after accounting for the impacts from stagnation and heat waves. The reason why the direction of the correlation changes after weighting can be either an increase or decrease is because the weights are assigned to both drought and non-drought months. The weighted enhancements of ozone are 30-59% lower than the original, un-weighted values, but remain to be significantly positive. The corresponding reduction for the $PM_{2.5}$ enhancements is 27%-45%. The west and southeast have a larger reduction in the enhancements of both pollutants after weighting, consistent with the fact that these regions show a larger increase in stagnation and heat wave frequencies during drought. The same weighting method can be separately applied to stagnation and heat waves to compare their effects individually (Figure S5-6). Stagnation exerts a larger influence on the weighted enhancement in the west and southeast, while heat waves has a larger effect in the Great Plains, consistent with the spatial distribution of their respective occurrences during drought. In all the cases examined here, the weighting does not change the sign or statistical significance of the SPEI-pollutant correlations at all the sites, indicating the covariance of drought with stagnation and heat waves might not be the dominant factor causing the SPEI-pollutant correlations. The weighting however reduces the magnitude of ozone and $PM_{2.5}$ enhancements associated with drought in every region, with an average reduction of 40% when both events are counted together as weights. This indicates that more frequent stagnation and heat waves could explain up to 40% of the ozone and $PM_{2.5}$ enhancements during drought, a significant but not majority factor.

3.3 Emission/deposition/chemistry factors for the drought-pollution association

~~Compared to normal (wet) conditions, the average enhancement during drought is 4.7 (7.9) ppb for O_3 and 2.4 (3.0) μg m^{-3} for $PM_{2.5}$ over the eastern sites. The relative change is consistent across different regions at about 7% for ozone and 18% for $PM_{2.5}$. The negative correlations as well as ozone/$PM_{2.5}$ enhancements under drought conditions using sc_PDSI_pm are similar to those derived with SPEI, thus validating the robustness of the response of ozone/$PM_{2.5}$ under drought conditions and suggesting little sensitivity of our analysis to the choice of drought indicator.~~

~~3.2 Causes of ozone and $PM_{2.5}$ enhancement by drought~~

Drought can further affect air quality through perturbations to emissions, deposition, and chemical processes ~~and deposition~~. High temperature conditions during drought will lead to higher production rate of ozone as well as higher emissions of BVOCs (Fuentes et al., 2000; Guenther et al., 2012).~~The ozone enhancement due to drought is primarily associated with higher temperatures (1.1℃ – 2.0℃) and stronger shortwave radiation (8.1-12.4 W m^{-2}) (Table S2). These conditions lead to higher production rate of ozone as well as higher emissions of BVOCs (Fuentes et al., 2000; Guenther et al., 2012).~~ Surface observations of isoprene suggest 7-20% higher concentrations under drought conditions (Table 1; Figure 4). An exception is a~~A~~ decrease in isoprene ~~is found~~ during severe drought (SPEI < -2) over the southeast and northeast US (Figure 4~~2~~), presumably due to shutoff of isoprene emissions when severe water stress causes reduction in carbon sources, lower level of isoprene synthase gene expression, stomata closure and wilting of vegetation (Pegoraro et al., 2004; Brilli et al., 2007; Seco et al., 2015). Surface NO_2 was

~~also~~ found to be higher by 0.07-1.26 ppb (2-9%) ~~higher~~, attributable to increased emissions from fires, soils and possibly the power sector. Precipitation scavenging of air pollutants should be much lower during drought, resulting in higher pollutant concentrations and longer lifetime in the atmosphere. Compiling the scattered measurements by the National Atmospheric Deposition Program, we found a 23-32% reduction in wet deposition of sulfate during drought (Table 1). In addition, severe drought can potentially lead to elevated surface ozone by reducing the ozone dry deposition to vegetation (Fowler et al., 2009; Kavassalis and Murphy, 2017). A modelling study suggested up to 20% reductions in ozone dry deposition due to lower stomatal conductance during a drought event in Texas (Huang et al., 2016). However, changes of wet and dry deposition fluxes due to drought are difficult to quantify due to dearth of deposition measurements.

~~In addition, ozone dry deposition can be reduced by up to 20% due to lower stomatal conductance under drought (Huang et al., 2016).~~

The enhancements of $PM_{2.5}$ species during drought are presented in Figure 5 at a subset of surface sites with speciation measurements. Organic aerosol (OA), sulfate and dust are major contributors to the overall $PM_{2.5}$ enhancements. There is a 2-15% increase in sulfate, attributable in part to reduced wet deposition.~~Changes in $PM_{2.5}$ species, presented in Figure 3 at a subset of surface sites with speciation measurements, indicate that organic aerosol (OA), sulfate and dust are major contributors to the overall $PM_{2.5}$ enhancements. Wet deposition of inorganic species measured by the National Atmospheric Deposition Program suggest a 23-32% reduction in sulfate wet deposition during drought (Table 1), explaining partly the 2-15% sulfate increase.~~ While oxidation rate of SO_2 ~~may~~ increase at high temperatures (Tai et al., 2010), ~~there is a~~surface SO_2 shows a 1-10% increase ~~in surface SO_2~~ during drought, presumably due to~~resulting from~~ reduced dry or wet deposition and higher emissions from fires and electricity generation (Scanlon et al., 2013). Dust enhancements are~~is~~ most significant in the west (27%) and the Great Plains (16%) due to more semi-arid areas. Significant OA enhancements (12-35%) are found associated with drought across all the US.~~Drought causes significant OA enhancements (12-35%) across all the US.~~ Fire emissions of primary OA are 1-3 times higher during drought, explaining a large portion of the OA enhancement (Tables S3-4). When excluding the fire influences, an increase in the OA to BC ratio was found under drought (Figure S8~~5~~), indicative of ~~suggesting~~ an increase in secondary organic aerosols (SOA) formation. However, routine networks provide only limited classification of OA and cannot fully distinguish the response of SOA to drought from that of total OA.

The above analysis suggests that the ozone and $PM_{2.5}$ enhancements during drought are largely responses of natural processes from the land biosphere and abnormal atmospheric conditions. ~~Changes in ozone and $PM_{2.5}$ are largely responses of natural processes from the land biosphere and abnormal atmospheric conditions.~~ To compare the drought~~-induced~~ related changes with the effects of anthropogenic emission reductions in the US, we divided the data into two sub-periods: 1990 to 2003 (P1) and 2004 to 2014 (P2). Anthropogenic emissions of $PM_{2.5}$ and ozone precursors have decreased significantly in the US from P1 to P2, for example, by 50% for SO_2 and 32% for NO_x according to the Air Pollutant Emission Trend Data from the US EPA (EPA, 2016). In spite of this, drought~~-induced~~ related enhancements of ozone and $PM_{2.5}$ ~~pollutants~~ are manifested clearly in ~~observational data for~~ both periods, ~~and~~ with little change in the magnitude of these enhancements ~~remains the same~~ between P1 and P2 (Table 2). Under normal conditions, ~~t~~there is a ~~he~~ decrease of ozone ~~(1.6 ppbv)~~ and $PM_{2.5}$ ~~(1.8 μg m^3)~~ from P1 to P2 by an average of1.6 ppbv and 1.8 μg m^3, respectively, which is attributable to the reductions of US anthropogenic emissions. ~~is attributable to the reductions of US anthropogenic emissions.~~ By comparison, drought~~-induced~~ related mean enhancement of ozone exceeds 4 ppbv in both periods and that of $PM_{2.5}$ is 1.6 μg m^3. Therefore, the pollutant enhancements associated with droughts do not appear to be affected

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by the decreasing trend of US anthropogenic emissions, indicating natural processes as the primary cause.

~~**This demonstrates the significantly adverse impact of increasing drought on air quality through natural processes, which offset the benefits of reductions in anthropogenic emissions.**~~

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3.~~4~~3 Modeled response of air pollutants to drought

Previous studies suggest that climate models have some skills to predict the variability of drought (Dai, 2012). Indeed the four models from ACCMIP all reproduce the observed spatial patterns of historical droughts in the US (Figure S7). Simulated severe droughts (model SPEI < -1.3) occur ~20% of the time over the west and southern US, consistent with the observed SPEI. However, the temporal correspondence (i.e. month-to-month) between model SPEI and observed 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 evaluation of simulated pollution responses to drought, because we used the model SPEI to derive the SPEI-pollutants relationships from each model.

~~Indeed the four models from ACCMIP all successfully reproduce the observed statistics of historical droughts in the US (Figure S6). In spite of this,~~ The models vary greatly in their ability of predicting the drought-pollutants relationships~~effects of drought on atmospheric composition~~. With respects to surface ozone, all the models are able to capture its negative correlation with SPEI over most of the US (Figure 6~~4~~), as they all predict some levels of increase in ozone production driven by higher temperatures during drought (Figure S9~~7~~). However, the simulated slopes and magnitude of ozone enhancement are less than half of the observed values in many regions, suggesting a lack of full representation of the drought effects~~, especially on natural emissions and dry deposition through drought perturbation of the land biosphere~~. The GISS-E2-R model, which is the only model that includes interactive isoprene emissions with model temperature, ~~which has BVOCs emissions coupled with weather/climate,~~ reproduces the observed isoprene increases. This allows the model to simulate ozone enhancements resulting from stronger isoprene emissions (Schnell et al., 2016), and thus the GISS model simulates the greater SPEI-ozone slope as compared to other models (Figure 6). In spite of lacking the interactive isoprene emissions, ~~and thus simulates relatively larger ozone enhancement under drought conditions (Figure S7). T~~The MICRO-CHEM model shows higher ozone enhancements than other models because it simulates the largest increase of ozone production caused by drought, presumably due to a larger sensitivity of ozone to temperature. Drought perturbation of the land biosphere would lead to reductions in the ozone dry deposition sink and hence higher ozone enhancements. For example, a model sensitivity study by Lin et al. (2017) showed that reducing ozone dry deposition velocity by 35% in the GFDL-AM3 model during the severe North American drought of 1988 would lead to 10 ppbv greater ozone enhancements than a simulation with constant dry deposition velocity. However, all the ACCMIP models examined here simulate little changes of ozone dry deposition (-3~5%) during drought.

~~All the models simulate little changes of ozone dry deposition (3~5%) during drought.~~

The models are less skillful in reproducing the effects of drought on $PM_{2.5}$ (Figure 7~~5~~). All the models incorrectly predict a decrease of $PM_{2.5}$ under drought conditions and hence a positive $PM_{2.5}$-SPEI relationship for many regions in the US, whereas this relationship is clearly negative in the observations across all the regions. For the few regions where some models are correct about the direction of the $PM_{2.5}$ change (e.g. the western US by GISS-E2-R and eastern US by NCAR-CAM3.5), the magnitude of the $PM_{2.5}$ change is less than 70% of that observed.

The model response is primarily driven by a ubiquitous and excessive decrease of sulfate under drought conditions caused by large reductions of sulfate production in clouds (-22~-73%) (Figure S10-11~~8 9~~). In contrast, only 14-34% of the sites in the west and the Great Plains show a decrease of sulfate during

drought. The model deficiency in sulfate can be explained by their underestimate of low-altitude cloud fraction at higher temperatures (Shen et al., 2016). This bias would lead to an underestimate of sulfate production as well as SOA-processing in clouds during drought (i.e. high temperature conditions), which could outweigh the aerosol deposition decrease. Figure 8 compares the satellite-derived sensitivity of total and boundary-layer CF to drought severity with that simulated by the GISS and GFDL model, which are the only ACCIMP models that archived layer-specific CF. For the boundary layer CF which should be more relevant for in-cloud processing of aerosols, the observed sensitivity averages about 0.51 per unit increase of SPEI, while the GISS and GFDL model shows a sensitivity of 4.37 and 3.41, respectively, about a factor of 8 higher than the observed value. The models also overestimate the sensitivity of total CF to drought, but to a less extent. Another important aspect of model deficiencies is that they all underestimate the OA enhancements in every region. Simulated OA changes primarily result from reduced wet deposition (~40%), lacking important contributions from changing BVOCs emissions, fires, or chemistry (Figure S10-11) as suggested by observations.

In summary,~~, which outweighs the deposition decrease. The simulated sensitivity of low-altitude cloud fraction to drought severity is 4.69 and 5.75 per unit increase of SPEI from the GISS and GFDL model respectively. By comparison, satellite-derived low-altitude cloud fraction shows a sensitivity of only 0.52 per unit increase of SPEI (Figure 6). In addition, the models underestimate the OA enhancements for all the regions. OA changes in the model are primarily resulted from reduced wet deposition (~40%), lacking important contributions from changing BVOCs emissions, fires, or chemistry (Figure S8-9).~~

t~~T~~he model deficiencies suggest a lack of mechanistic understanding of natural processes of importance to atmospheric composition and/or their perturbations by drought, although attribution of the underlying causes would require chemistry-climate model sensitivity experiments, which is outside the scope of the present study. Emissions, deposition, and chemistry are the most important aspects of model configurations affecting the drought-pollutants relationship. Since natural emissions were not specified, the ACCIMP models treated natural emissions differently, which is a key factor in the different performance between models. Using the observed SPEI-pollutants relationship as a diagnostic, we found that the model with interactive isoprene emissions (e.g. the GISS model) has a better ability to simulate the SPEI-ozone relationship, indicating the importance of drought effects on BVOCs emissions. With regard to deposition, all the models simulate some levels of decreasing wet deposition during drought, but dry deposition is largely insensitive to drought due to the lack of drought effects on the properties of the land and biosphere. The overestimate of the dry deposition sink during drought may be another reason behind the models' deficiency in underestimating the drought-pollutants relationship. Lastly, all the models overestimate the sulfate reduction, but at the same time underestimate the OA increase during drought. Both problems might be caused by the model misrepresentation of cloud sensitivity to changing drought severity, although the OA bias could also be caused by uncertainties of fire and BVOC emissions in the model. ~~drought effects on natural processes of importance to atmospheric composition. Specifically, the lack of biosphere responses and misrepresentation of cloud sensitivity to changing drought severity are the major contributors of the model deficiencies. While all the models simulate some levels of decreasing wet deposition, dry deposition in the model is largely insensitive to drought due to the lack of drought effects on the properties of the land and biosphere. Most models do not have the effects of drought on different types of natural emissions, e.g. from fires, BVOCs, and dust.~~

4. Future changes in drought and adverse impacts on air quality

To circumvent the model deficiencies, the effects of future increases of drought on air quality were estimated by extrapolating their present-day relationships from observations to model projected drought occurrences under future warming scenarios. Projected changes in SPEI from the present to future

climate were derived from the outputs of the four ACCMIP models (i.e. GISS-E2-R, GFDL-CM3, CCSM4, and MIROC-ESM-CHEM) archived by the Coupled Model Intercomparison Project Phase 5 (CMIP5) (Taylor et al., 2012). The CMIP5 historical runs cover the period from 1850 to near present, and are forced with observed changes in atmospheric composition with evolving land cover. The future projection runs span from 2006 to 2300, forced with specified concentrations of certain atmospheric constituents defined in three representative concentration pathways (RCPs) scenarios (Moss et al., 2010): RCP 2.6 (low mitigation emission scenario), RCP 4.5 (midrange mitigation emission scenario) and RCP 8.5 (high emission scenario). Changes in future drought conditions compared to the present are defined as the 2100 SPEI (2080-2099 mean) minus its value in 2000 (1990-2005 mean).

Figure 9~~7~~a shows the projection of SPEI in the US by 2100 (2080-2099 mean) under different RCPs that are derived from the mean of the four models from the CMIP5 outputs. Drought risks are projected to increase with warming scenarios over all parts of the US, with the largest increases in the west and the Great Plains, consistent with previous projections (Cook et al., 2015). These projected SPEI changes (2100 minus 2000), when multiplied by the present-day relationship between SPEI and air quality derived from observations (c.f. Figure 1), suggests a 0.3-3.0 ppb (1-6%) increase of surface ozone and 0.1-1.0 μg m^{-3} (1-16%) increase of $PM_{2.5}$ in the US in 2100 as a result of increasing drought alone under different RCPs (Figure 7b-c). The increase of ozone and $PM_{2.5}$ are largest in the west. The maximum increase is 14% for ozone and 41% for $PM_{2.5}$ under the extreme warming scenario (RCP 8.5), significantly higher than the present-day effects. While this extrapolation-based projection may not be reliable quantitatively, it suggests a significant climate change penalty on air quality through drought, which has been overlooked before and pose a new challenge for air quality managers.

5. Discussion

The retrospective analysis of observations demonstrates that past droughts have ~~caused~~ been associated with significant deterioration of air quality through natural processes, resulting in potentially large tolls on public health that have not been considered in previous impact analysis of drought. The land biosphere plays a key role in mediating drought-~~induced~~ related changes in atmospheric chemistry. The magnitude of the land biosphere response is largely dependent on concurrent changes in solar irradiance, temperature and water at different levels of drought severity and duration. More sunlight and higher temperatures may outweigh some levels of water stress, resulting in enhanced BVOCs emissions through leaf biochemistry, vapor pressure difference and underlying metabolism processes (Fuentes et al., 2000). However, extreme and/or prolonged drought conditions with severe water stress coupled with very high temperatures can affect the activity of enzyme and health of the plants, therefore leading to reductions in BVOCs emissions. More comprehensive understanding of the land biosphere responses is required to quantify the impact of land biosphere to atmospheric compositions under different drought conditions. In addition to changing BVOCs emissions, reduced aerosol water content under drought conditions can perturb aqueous phase formation of SOA from BVOCs, but the impact is not clear (Gilardoni et al., 2016). Changes in anthropogenic emissions under drought conditions are also uncertain. Local land use type and water management policy can significantly affect human reactions to drought. Furthermore, the interaction between anthropogenic emissions and natural responses further compound the drought effect, as anthropogenic emitted gases and aerosols can affect the oxidation and partitioning processes of SOA from BVOCs (Hoyle et al., 2011; Xu et al., 2015).

Changes in the land biosphere and atmospheric compositions, including gases and aerosols, can provide feedbacks to the climate through radiative effects and cloud interactions. Reductions in vegetation cover affect surface albedo and dust emissions, resulting in enhanced surface temperatures, intensification of drought conditions and geographical shift of drought pattern (Cook et al., 2009). Increasing wildfire

activity and fire-emitted aerosols alter the regional energy budget and circulation, which lead to reduced precipitation thus further enhancing drought severity and vulnerability of ecosystem towards wildfires (Bevan et al. 2009; Tosca et al., 2010; Hodnebrog et al., 2016). Improvements in climate-chemistry models are thus imperative to facilitate better prediction of atmospheric composition changes due to changes in drought and improved understanding of the associated feedbacks of composition changes to climate and drought itself.

The observational analysis presented here indicates significant changes of air pollutants under drought conditions. However, it is not sufficient to quantify the full extent of the cascading effects of drought on the complex chemistry of ozone and SOA, which would require more targeted measurements providing for example more classification of organic materials and modeling at the process level. Uncertainties exist in the model assessment since we are using a single version of simulation for each model and the study period is relatively short and may not represent the full simulation results. Nonetheless, both observations and model indicate the important role of the land biosphere and atmospheric conditions in regulating pollutant levels under drought conditions. Future air quality management should consider the adverse effects from increasing drought risks.

Data availability

All datasets used in this study are publically accessible.

Author contribution

Y. W. and Y. X. conceived the research idea. Y. X. and W.D. performed the analysis and Y. W. wrote the initial draft of the paper. All authors contributed to the interpretation of the results and the preparation of the manuscript.

Competing financial interests

The authors declare no competing financial interests.

Acknowledgements

We thank the individuals and groups involved in making observations at IMPROVE, US EPA, CASNET, NADP and PAMS networks, and in preparing the SPEI, PDSI, GFED, ASI, ~~MERRA~~ CRU, ERA-Interim and ISCCP-D1~~2~~like database. We thank the modelling groups that participate in ACCMIP and CMIP5 for producing the model outputs and making them available. This work was supported in part by the National Key Basic Research Program of China (2013CB956603 and 2014CB441302). J. Wang acknowledges the support from NASA Aura Science program (grant #: NNX14AG01G managed by Dr. Ken Jucks), Applied Science Program (grant #: NNX15AC28A managed by Dr. John Haynes), and ACMAP program (grant #: NNX15AC30G managed by Dr. Richard Eckman).

References

Akagi, S., Yokelson, R. J., Wiedinmyer, C., Alvarado, M., Reid, J., Karl, T., Crounse, J., and Wennberg, P.: Emission factors for open and domestic biomass burning for use in atmospheric models, *Atmospheric Chemistry and Physics*, 11, 4039-4072, 2011.

Allen, R. J., Landuyt, W., and Rumbold, S. T.: An increase in aerosol burden and radiative effects in a warmer world, *Nature Climate Change*, 6, 269-274, 10.1038/nclimate2827, 2015.

Arnell, N. W.: Climate change and global water resources: SRES emissions and socio-economic scenarios, *Global environmental change*, 14, 31-52, 2004.

Beguer á, S., Vicente-Serrano, S.M., Reig, F. and Latorre, B.: Standardized precipitation evapotranspiration index (SPEI) revisited: parameter fitting, evapotranspiration models, tools, datasets and drought monitoring. International Journal of Climatology, 34(10), pp.3001-3023, 2004.

Bevan, Suzanne L., et al.: Impact of atmospheric aerosol from biomass burning on Amazon dry‐season drought. *Journal of Geophysical Research: Atmospheres* 114.D9, 2009.

Brilli, F., Barta, C., Fortunati, A., Lerdau, M., Loreto, F., and Centritto, M.: Response of isoprene emission and carbon metabolism to drought in white poplar (Populus alba) saplings, *New Phytologist*, 175, 244-254, 2007.

Cook, B. I., Miller, R. L., and Seager, R.: Amplification of the North American "Dust Bowl" drought through human-induced land degradation, *Proceedings of the National Academy of Sciences*, 106, 4997-5001, 2009.

Cook, B. I., Ault, T. R., and Smerdon, J. E.: Unprecedented 21st century drought risk in the American Southwest and Central Plains, *Science Advances*, 1, e1400082, 2015.

Dai, A.: Characteristics and trends in various forms of the Palmer Drought Severity Index during 1900–2008, *Journal of Geophysical Research: Atmospheres*, 116, 2011.

Dai, A.: Increasing drought under global warming in observations and models, *Nature Climate Change*, 3, 52-58, 10.1038/nclimate1633, 2012.

Davidson, E. A., Nepstad, D. C., Ishida, F. Y., and Brando, P. M.: Effects of an experimental drought and recovery on soil emissions of carbon dioxide, methane, nitrous oxide, and nitric oxide in a moist tropical forest, *Global Change Biology*, 14, 2582-2590, 2008.

Dawson, J., Adams, P., and Pandis, S.: Sensitivity of PM 2.5 to climate in the Eastern US: a modeling case study, *Atmospheric Chemistry and Physics*, 7, 4295-4309, 2007.

Dee, D. P. et al. The ERA-Interim reanalysis: configuration and performance of the data assimilation system, *Quart. J. Roy. Meteorol. Soc.*, 137, 553–597 (2011).

Filleul, L., Cassadou, S., M édina, S., Fabres, P., Lefranc, A., Eilstein, D., Le Tertre, A., Pascal, L., Chardon, B., Blanchard, M., Declercq, C., Jusot, J.-F., Prouvost, H., and Ledrans, M.: The Relation Between Temperature, Ozone, and Mortality in Nine French Cities During the Heat Wave of 2003, *Environmental health perspectives*, 114, 1344-1347, 10.1289/ehp.8328, 2006.

Fowler D, Pilegaard K, Sutton M, Ambus P, Raivonen M, Duyzer J, *et al.* Atmospheric composition change: ecosystems–atmosphere interactions. *Atmospheric Environment* 2009, **43**(33)**:** 5193-5267.

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Forouzanfar, M. H., Alexander, L., Anderson, H. R., Bachman, V. F., Biryukov, S., Brauer, M., Burnett, R., Casey, D., Coates, M. M., and Cohen, A.: Global, regional, and national comparative risk assessment of 79 behavioural, environmental and occupational, and metabolic risks or clusters of risks in 188 countries, 1990–2013: a systematic analysis for the Global Burden of Disease Study 2013, *The Lancet*, 386, 2287-2323, 2015.

Fuentes, J. D., Gu, L., Lerdau, M., Atkinson, R., Baldocchi, D., Bottenheim, J., Ciccioli, P., Lamb, B., Geron, C., and Guenther, A.: Biogenic hydrocarbons in the atmospheric boundary layer: a review, *Bulletin of the American Meteorological Society*, 81, 1537-1575, 2000.

Giglio, L., Randerson, J. T., and Werf, G. R.: Analysis of daily, monthly, and annual burned area using the fourth - generation global fire emissions database (GFED4), *Journal of Geophysical Research: Biogeosciences*, 118, 317-328, 2013.

Gilardoni, S., Massoli, P., Paglione, M., Giulianelli, L., Carbone, C., Rinaldi, M., Decesari, S., Sandrini, S., Costabile, F., and Gobbi, G. P.: Direct observation of aqueous secondary organic aerosol from biomass-burning emissions, *Proceedings of the National Academy of Sciences*, 113, 10013-10018, 2016.

Griffin, D., and Anchukaitis, K. J.: How unusual is the 2012–2014 California drought?, *Geophysical Research Letters*, 41, 9017-9023, 2014.

Guenther, A. B., et al.: The Model of Emissions of Gases and Aerosols from Nature version 2.1 (MEGAN2. 1): an extended and updated framework for modeling biogenic emissions. *Geoscientific Model Development*, 5.6: 1471-1492, 2012.

Guenther, A. B.: Bidirectional exchange of volatile organic compounds.*Review and Integration of Biosphere-Atmosphere Modelling of Reactive Trace Gases and Volatile Aerosols*. Springer Netherlands, 107-113, 2015.

Heim Jr and Richard R.: A review of twentieth-century drought indices used in the United States. *Bulletin of the American Meteorological Society*, 83.8,1149-1165, 2002.

Hodnebrog, Ø., Myhre, G., Forster, P. M., Sillmann, J., and Samset, B. H.: Local biomass burning is a dominant cause of the observed precipitation reduction in southern Africa, *Nature communications*, 7, 2016.

Hoerling, M., Eischeid, J., Kumar, A., Leung, R., Mariotti, A., Mo, K., Schubert, S., and Seager, R.: Causes and predictability of the 2012 Great Plains drought, *Bulletin of the American Meteorological Society*, 95, 269-282, 2014.

Hou, P., and Wu, S.: Long-term Changes in Extreme Air Pollution Meteorology and the Implications for Air Quality, *Scientific reports*, 6, 2016.

Hoyle, C., Boy, M., Donahue, N., Fry, J., Glasius, M., Guenther, A., Hallar, A., Huff Hartz, K., Petters, M., and Petäjä, T.: A review of the anthropogenic influence on biogenic secondary organic aerosol, *Atmospheric Chemistry and Physics*, 11, 321-343, 2011.

Huang, L., McDonald-Buller, E. C., McGaughey, G., Kimura, Y., and Allen, D. T.: The impact of drought on ozone dry deposition over eastern Texas, *Atmospheric Environment*, 127, 176-186, 10.1016/j.atmosenv.2015.12.022, 2016.

Kavassalis, S.C. and Murphy, J.G.: Understanding ozone-meteorology correlations: A role for dry deposition. Geophysical Research Letters, 44(6), pp.2922-2931, 2017

Lamarque, J.-F., Bond, T. C., Eyring, V., Granier, C., Heil, A., Klimont, Z., Lee, D., Liousse, C., Mieville, A., and Owen, B.: Historical (1850–2000) gridded anthropogenic and biomass burning emissions of reactive gases and aerosols: methodology and application, *Atmospheric Chemistry and Physics*, 10, 7017-7039, 2010.

Lamarque, J., Shindell, D. T., Josse, B., Young, P., Cionni, I., Eyring, V., Bergmann, D., Cameron-Smith, P., Collins, W. J., and Doherty, R.: The Atmospheric Chemistry and Climate Model Intercomparison Project (ACCMIP): overview and description of models, simulations and climate diagnostics, *Geoscientific Model Development*, 6, 179-206, 2013.

Lelieveld, J., Evans, J. S., Fnais, M., Giannadaki, D., and Pozzer, A.: The contribution of outdoor air pollution sources to premature mortality on a global scale, Nature, 525, 367-371, 10.1038/nature15371, 2015.

Lin M ,Larry W, Arlene M,. US surface ozone trends and extremes from 1980 to 2014: quantifying the roles of rising Asian emissions, domestic controls, wildfires, and climate. *Atmos Chem Phys* 2017, **1680:** 7324.

Malm, W. C., Sisler, J. F., Huffman, D., Eldred, R. A., and Cahill, T. A.: Spatial and seasonal trends in particle concentration and optical extinction in the United States, *Journal of Geophysical Research: Atmospheres*, 99, 1347-1370, 1994.

Minnis, Patrick, *et al*. : CERES edition-2 cloud property retrievals using TRMM VIRS and Terra and Aqua MODIS data—Part I: Algorithms. *IEEE Transactions on Geoscience and Remote Sensing* 49.11: 4374-4400, 2011.

Moss, R. H., Edmonds, J. A., Hibbard, K. A., Manning, M. R., Rose, S. K., Van Vuuren, D. P., Carter, T. R., Emori, S., Kainuma, M., and Kram, T.: The next generation of scenarios for climate change research and assessment, *Nature*, 463, 747-756, 2010.

Nielsen-Gammon, J. W.: The 2011 Texas drought, *Texas Water Journal*, 3, 59-95, 2012.

Pegoraro, E., Rey, A., Greenberg, J., Harley, P., Grace, J., Malhi, Y., and Guenther, A.: Effect of drought on isoprene emission rates from leaves of Quercus virginiana Mill, *Atmospheric Environment*, 38, 6149-6156, 2004.

Prospero, J. M., and Lamb, P. J.: African droughts and dust transport to the Caribbean: Climate change implications, *Science*, 302, 1024-1027, 2003.

Qu, W., Wang, J., Zhang, X., Yang, Z., and Gao, S.: Effect of cold wave on winter visibility over eastern China, *Journal of Geophysical Research: Atmospheres*, 120, 2394-2406, 2015.

Randerson, J., Chen, Y., Werf, G., Rogers, B., and Morton, D.: Global burned area and biomass burning emissions from small fires, *Journal of Geophysical Research: Biogeosciences*, 117, 2012.

Rienecker, M. M., Suarez, M. J., Gelaro, R., Todling, R., Bacmeister, J., Liu, E., Bosilovich, M. G., Schubert, S. D., Takacs, L., and Kim, G.-K.: MERRA: NASA's modern-era retrospective analysis for research and applications, *Journal of Climate*, 24, 3624-3648, 2011.

Rosenzweig, C., Iglesias, A., Yang, X., Epstein, P. R., and Chivian, E.: Climate change and extreme weather events; implications for food production, plant diseases, and pests, *Global change & human health*, 2, 90-104, 2001.

Seco, R., Karl, T., Guenther, A., Hosman, K.P., Pallardy, S.G., Gu, L., Geron, C., Harley, P. and Kim, S.: Ecosystem‐scale volatile organic compound fluxes during an extreme drought in a broadleaf

temperate forest of the Missouri Ozarks (central USA). *Global change biology*, 21.10: 3657-3674, 2015.

Scanlon, B. R., Duncan, I., and Reedy, R. C.: Drought and the water–energy nexus in Texas, *Environmental Research Letters*, 8, 045033, 2013.

Schnell JL, Prather MJ, Josse B, Naik V, Horowitz LW, Zeng G, *et al.* Effect of climate change on surface ozone over North America, Europe, and East Asia. *Geophysical Research Letters*, 43(7)**:** 3509-3518, 2016

Schnell, J.L. and Prather, M.J.: Co-occurrence of extremes in surface ozone, particulate matter, and temperature over eastern North America. Proceedings of the National Academy of Sciences, 114(11), pp.2854-2859, 2016

Shen, L., Mickley, L. J., and Murray, L. T.: Strong influence of 2000–2050 climate change on particulate matter in the United States: Results from a new statistical model, *Atmos. Chem. Phys. Discuss*., doi:10.5194, 2016.

Steiner, A. L., Davis, A. J., Sillman, S., Owen, R. C., Michalak, A. M., and Fiore, A. M.: Observed suppression of ozone formation at extremely high temperatures due to chemical and biophysical feedbacks, *Proceedings of the National Academy of Sciences*, 107, 19685-19690, 2010.

Stephens, Graeme L., Norman B. Wood, and Philip M. Gabriel: An assessment of the parameterization of subgrid-scale cloud effects on radiative transfer. Part I: Vertical overlap. *Journal of the atmospheric sciences* 61.6: 715-732 2004.

Tai, A. P. K., Mickley, L. J., and Jacob, D. J.: Correlations between fine particulate matter (PM2.5) and meteorological variables in the United States: Implications for the sensitivity of PM2.5 to climate change, *Atmospheric Environment*, 44, 3976-3984, 10.1016/j.atmosenv.2010.06.060, 2010.

Taylor, K. E., Stouffer, R. J., and Meehl, G. A.: An overview of CMIP5 and the experiment design, *Bulletin of the American Meteorological Society*, 93, 485, 2012.

Tian, H., Ren, W., Tao, B., Sun, G., Chappelka, A., Wang, X., Pan, S., Yang, J., Liu, J., and S Felzer, B.: Climate extremes and ozone pollution: a growing threat to China's food security, Ecosystem Health and Sustainability, 2, 2016.

Tosca, M., Randerson, J., Zender, C., Flanner, M., and Rasch, P. J.: Do biomass burning aerosols intensify drought in equatorial Asia during El Niño?, *Atmospheric Chemistry and Physics*, 10, 3515-3528, 2010.

Trenberth, K. E., Dai, A., van der Schrier, G., Jones, P. D., Barichivich, J., Briffa, K. R., and Sheffield, J.: Global warming and changes in drought, *Nature Climate Change*, 4, 17-22, 2014.

University of East Anglia Climatic Research Unit; Harris, I.C.; Jones, P.D. (2014): CRU TS3.22: Climatic Research Unit (CRU) Time-Series (TS) Version 3.22 of High Resolution Gridded Data of Month-by-month Variation in Climate (Jan. 1901- Dec. 2013). NCAS British Atmospheric Data Centre, 24 September 2014. doi:10.5285/18BE23F8-D252-482D-8AF9-5D6A2D40990C.

U.S. Environmental Protection Agency: Technology Transfer Network Clearinghouse for Inventories and Emission Factors, National Emissions Inventory (NEI) air pollutant emissions trendsdata, 2016, https://www.epa.gov/air-emissions-inventories/air-pollutant-emissions-trends-data (last access: 10 Feb 2017).

Van der Werf, G. R., Randerson, J. T., Giglio, L., Collatz, G., Mu, M., Kasibhatla, P. S., Morton, D. C., DeFries, R., Jin, Y. v., and van Leeuwen, T. T.: Global fire emissions and the contribution of deforestation, savanna, forest, agricultural, and peat fires (1997–2009), *Atmospheric Chemistry and Physics*, 10, 11707-11735, 2010.

Vicente-Serrano, S. M., Beguer á, S., and L ópez-Moreno, J. I.: A Multiscalar Drought Index Sensitive to Global Warming: The Standardized Precipitation Evapotranspiration Index, *Journal of Climate*, 23, 1696-1718, 10.1175/2009jcli2909.1, 2010.

Wang, Y., Xie, Y., Cai, L., Dong, W., Zhang, Q., and Zhang, L.: Impact of the 2011 southern us drought on ground-level Fine aerosol concentration in summertime*, *Journal of the Atmospheric Sciences*, 72, 1075-1093, 2015.

Westerling, A. L., Gershunov, A., Brown, T. J., Cayan, D. R., and Dettinger, M. D.: Climate and wildfire in the western United States, Bulletin of the American Meteorological Society, 84, 595-604, 2003.

Westerling, A. L., and Swetnam, T. W.: Interannual to decadal drought and wildfire in the western United States, EOS, Transactions American Geophysical Union, 84, 545-555, 2003b

Xu, L., Guo, H., Boyd, C. M., Klein, M., Bougiatioti, A., Cerully, K. M., Hite, J. R., Isaacman-VanWertz, G., Kreisberg, N. M., and Knote, C.: Effects of anthropogenic emissions on aerosol formation from isoprene and monoterpenes in the southeastern United States, *Proceedings of the National Academy of Sciences*, 112, 37-42, 2015.

Zhang, H., Y. Wang, T.-W. Park, and Y. Deng, Quantifying the relationship between extreme air pollution events and extreme weather events, Atmos. Res., 188, 64-79, doi:10.1016/j.atmosres.2016.11.010, 2017.

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

Tables

Table 1: Changes in the concentrations of atmospheric gaseous compositions and sulfate wet deposition under drought compared to normal conditions. Data are from different measurement networks (see Method).

		West	Great Plains	Southeast	Northeast
NO_2 (ppb)	N[a]	130	27	81	122
	Diff	1.26 (+9.0%)	0.07 (+2.3%)	0.14 (+2.6%)	0.46 (+3.9%)
	p-value[b]	<0.01	0.68	0.25	<0.01
SO_2 (ppb)	N	66	28	113	290
	Diff	0.14 (+2.6%)	0.13 (+1.4%)	0.29 (+10.4%)	0.32 (+7.3%)
	p-value	0.05	0.28	<0.01	<0.01
Isoprene (ppb carbon)	N	8	14	28	21
	Diff	0.21 (+11.6%)	0.01 (+7.0%)	0.09 (+13.8%)	0.36 (+19.5%)
	p-value	0.04	0.60	0.09	0.01
Sulfate wet deposition (kg $month^{-1}$)	N	48	30	47	83
	Diff	-0.62 (-31.7%)	-1.39 (-26.7%)	-2.47 (-22.9%)	-2.99 (-26.2%)
	p-value	<0.01	<0.01	<0.01	<0.01

a. Number of sites.
b. P value derived from student t-test.

Table 2: Changes in the concentrations of ozone and $PM_{2.5}$ at two periods under drought compared to normal conditions.

	P1 (1990-2003)			P2 (2004-2014)			P2 minus P1
	Drought	**Normal**	**Diff**	**Drought**	**Normal**	**Diff**	**Normal**
				Ozone (ppbv)			
West	56.61	51.58	5.03	53.19	49.15	4.04	2.43
Great Plains	51.64	47.23	4.41	52.23	47.75	4.48	-0.52
Southeast	51.98	47.01	4.97	49.03	44.75	4.28	2.26
Northeast	51.64	46.43	5.21	48.19	44.23	3.96	2.20
Average	52.97	48.06	4.91	50.66	46.47	4.19	1.59
				$PM_{2.5}$ ($\mu g\ m^3$)			
West	6.57	5.56	1.01	5.84	4.74	1.10	0.82
Great Plains	7.69	6.22	1.47	6.86	5.81	1.05	0.41
Southeast	15.98	14.19	1.79	13.79	11.45	2.34	2.74
Northeast	16.37	14.25	2.12	12.67	10.94	1.73	3.31
Average	11.65	10.06	1.60	9.79	8.24	1.56	1.82

Figures

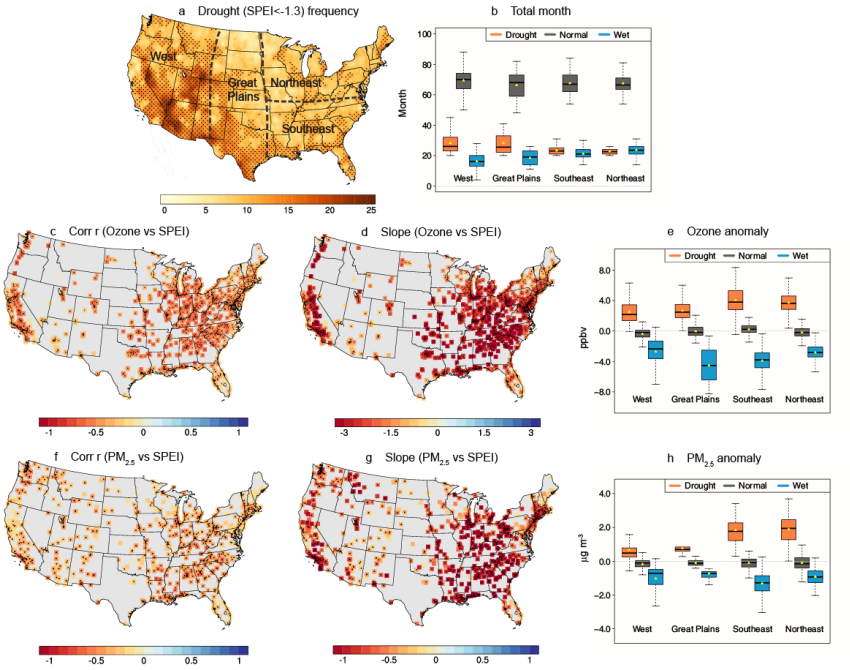

Figure 1. (a) Percentage occurrence of severe drought months (SPEI < -1.3) over the continental US during 1990-2014; black dots indicate drought frequency greater than 10% and dashed lines show the four geographical regions. Linear regression correlation coefficient r and slope of SPEI with O_3 (c,d) and $PM_{2.5}$ (f,g) anomalies at surface sites with data records longer than 5 years; yellow dots indicate regression significance at 95% confidence level. Boxplot comparisons of total months in different dry sectors (b), ozone (e) and $PM_{2.5}$ anomalies (h) under drought (SPEI < -1.3), normal (-0.5< SPEI < 0.5) and wet conditions (SPEI > 1.3) by region; the yellow triangles in the boxplot indicate mean values. All the surface data shown in the boxplot are restricted to sites with data records longer than 5 years and more than 10% occurrence of severe drought (SPEI < -1.3).

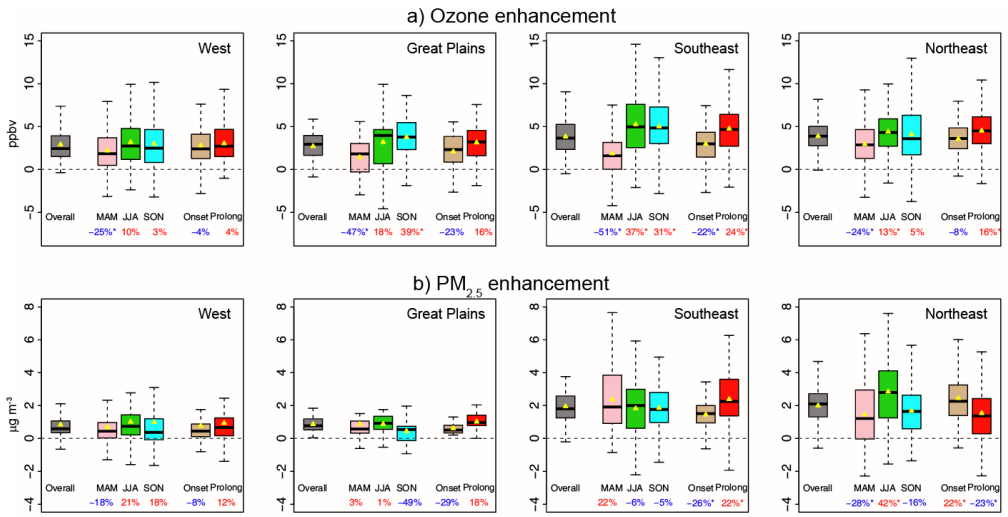

Figure 2. Ozone and $PM_{2.5}$ enhancements during drought relative to normal conditions at different seasons and different drought stages. The yellow triangles in the boxplot indicate mean values. The numbers below each box represent the difference relative to the overall enhancement of the whole growing season (Mar-Oct) (grey box), with the asterisks indicating significant differences at 95% level from the Student's t-test

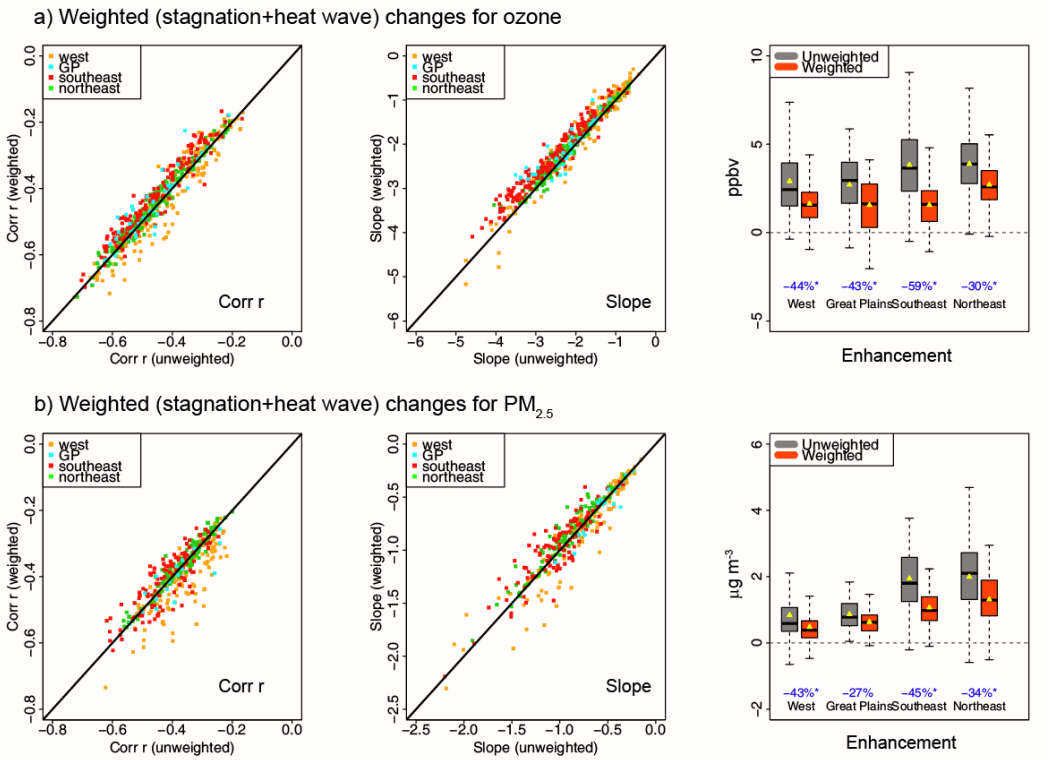

Figure 3. Comparison between the weighted (by frequency of stagnation and heat waves) and un-weighted SPEI-pollutants relationship (correlation r, left panel; correlation slope, right panel) and pollutants enhancements (right panel). The upper panel is for ozone and the lower panel for $PM_{2.5}$. Left and middle panels: the black lines are the 1:1 lines and different colors represent different regions. Right panel: the numbers below each box indicate the difference relative to the un-weighted enhancements.

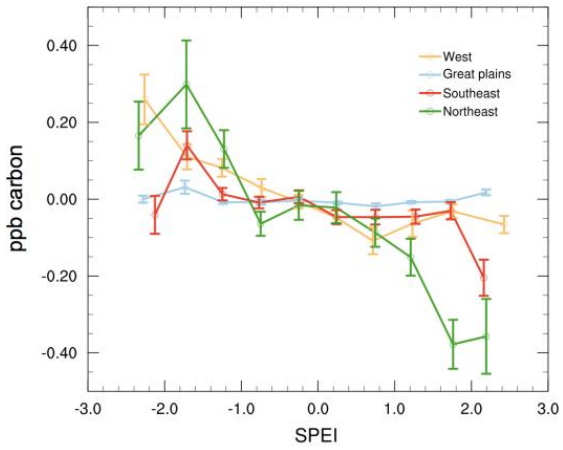

Figure 42: Isoprene anomalies (ppb carbon) derived from the PAMS network at binned SPEI levels over the Western, the Great Plains, the Southeastern and Northeastern US. Error bars indicate standard error of the mean.

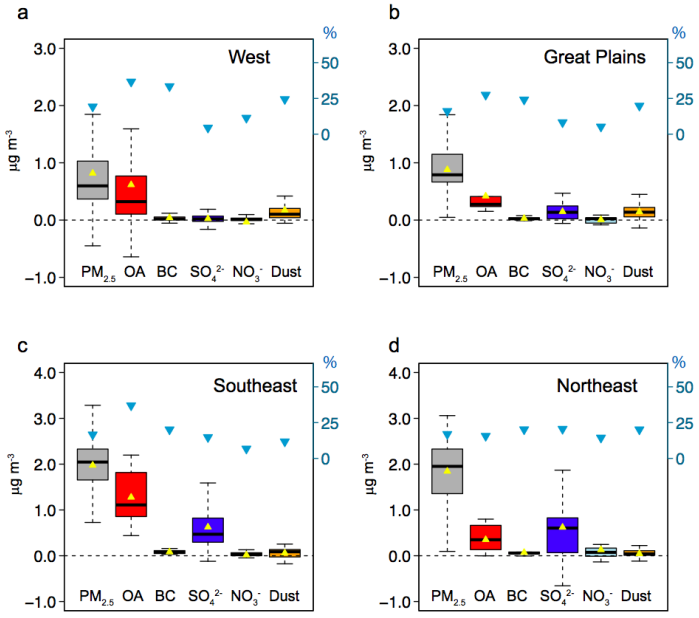

Figure 53: Boxplot of anomalies in $PM_{2.5}$ speciation during drought (SPEI < -1.3) compared to normal (-0.5 < SPEI< 0.5) conditions for the Western (a), Great Plains (b), Southeastern (c) and Northeastern (d) US. The yellow triangle indicates mean values and blue triangles indicate relative changes (right y-axis).

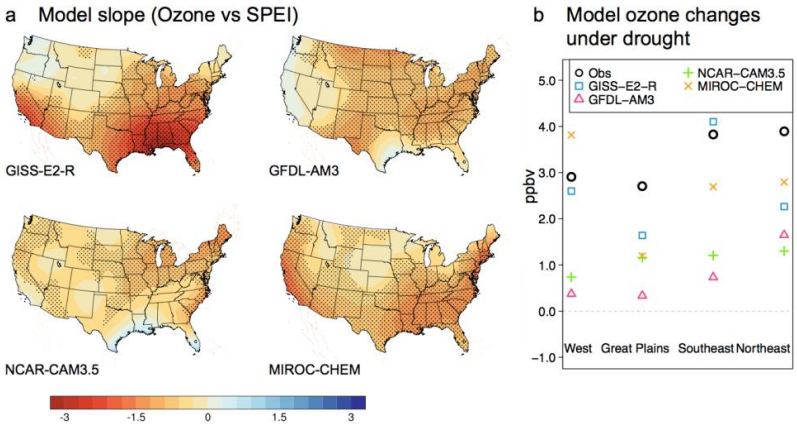

Figure 64: Linear regression slope between model derived SPEI and simulated ozone from GISS-E2-R, GFDL-AM3, NCAR-CAM3.5 and MIROC-CHEM model (a). Black dots represent regression significance at 95% confidence level. Note the color bar of (a) is the same as in Figure 1c. Comparison for the observed (black circle) and simulated changes (colored symbols) in ozone (b) under drought (SPEI < -1.3) compared to normal (-0.5 < SPEI < 0.5) condition by region.

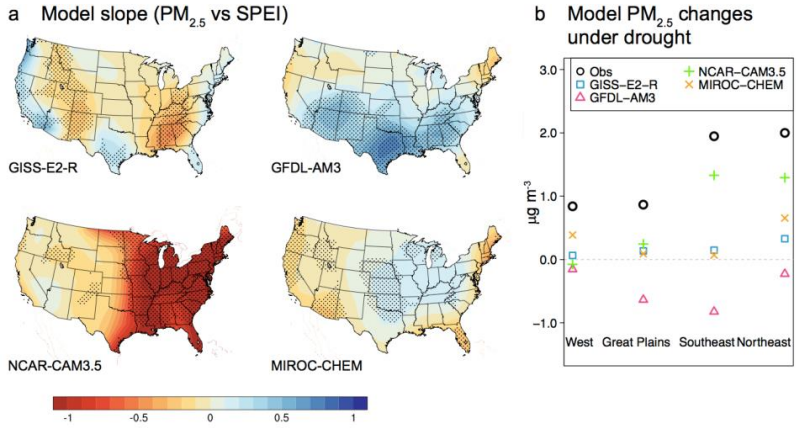

Figure 75: Linear regression slope between model derived SPEI and simulated $PM_{2.5}$ from GISS-E2-R, GFDL-AM3, NCAR-CAM3.5 and MIROC-CHEM model (a). Black dots represent regression significance at 95% confidence level. Note the color bar of (a) is the same as in Figure 1e. Comparison for the observed (black circle) and simulated changes (colored symbols) in $PM_{2.5}$ (b) under drought (SPEI < -1.3) compared to normal (-0.5 < SPEI < 0.5) condition by region.

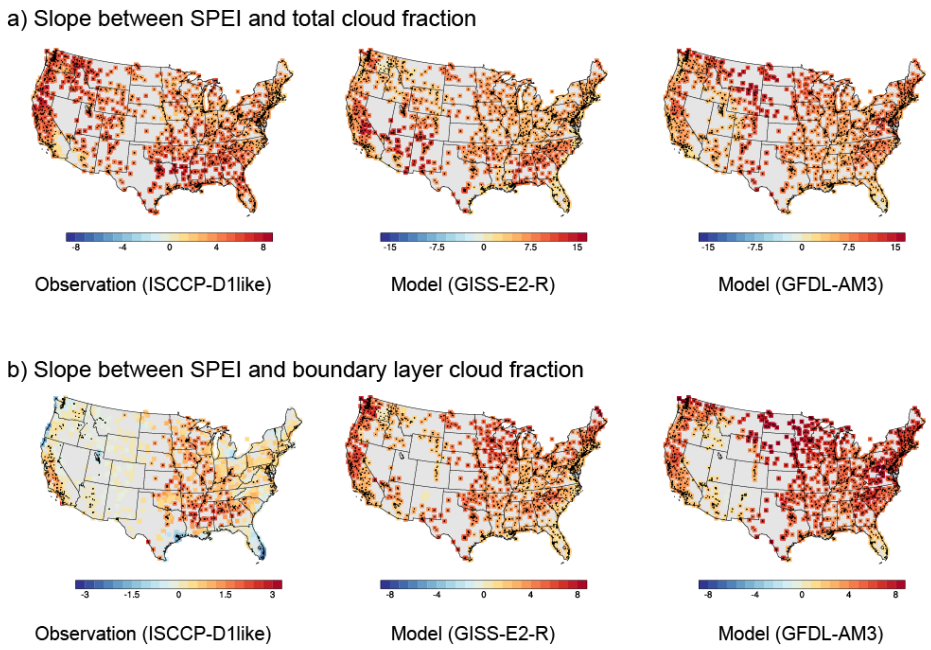

Figure 8. Slopes from linear regression between SPEI and (a) total and (b) boundary layer cloud fractions from the ISCCP satellite observations (left), GISS-E2-R (middle) and GFDL-AM3 model (right)

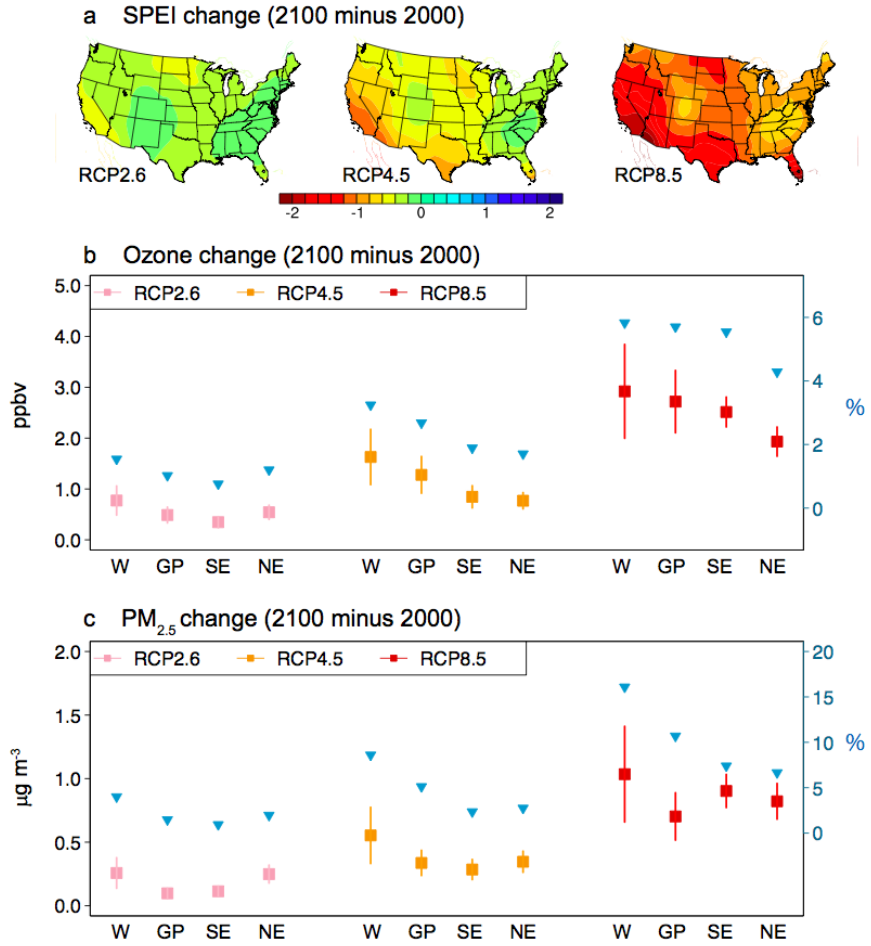

Figure 9: Predicted changes in SPEI between 2100 (2080-2099 average) and 2000 (1990-2005 average) by region under three RCP scenarios (a) from mean of four models (GISS-E2-R, GFDL-CM3, CCSM4 and MIROC-ESM-CHEM) and the estimated changes in surface ozone (b) and $PM_{2.5}$ (c) resulting from the SPEI changes alone. The four points in each RCP scenario represent the Western, Great Plains, Southeastern and Northeastern US. Error bar represents 1/2 standard deviation. Blue triangles indicate the mean percentage changes relative to the 2000 conditions (right y-axis).