**Supplementary materials**

**Table S1: Technical details for four ACCMIP models**

| | GISS-E2-R | GFDL-AM3 | NCAR-CAM3.5 | MIROC-CHEM |
|---|---|---|---|---|
| Available model output for historical runs | 1981-2005 | 1981-1990, 2001-2010 | 1982-1989, 2002-2009 | 1980-1984, 2000-2010 |
| Emissions — Anthropogenic | Prescribed | | | |
| Emissions — Biomass | Prescribed by decadal means | | | |
| Emissions — Biogenic | Interactive | Prescribed | | |
| Detrend & deseasonalize method | 7-yr moving average | Subtract time slice average by month | Subtract time slice average by month | Subtract time slice average by month |

10 **Table S2: Changes in meteorological variables at surface sites under drought conditions from the MERRA reanalysis**

| Diff * (drought minus normal) | West | Great Plains | Southeast | Northeast |
|---|---|---|---|---|
| Temp (℃) | 1.98 | 1.85 | 1.06 | 1.11 |
| Precipitation (mm month$^{-1}$) | -14.14 (-42.2%) | -23.87 (-44.6%) | -35.30 (-33.0%) | -27.40 (-56.4%) |
| Cloud fraction | -0.033 (-34.6%) | -0.027 (-39.2%) | -0.037 (-25.9%) | -0.043 (-31.7%) |
| Shortwave Radiation (W m$^{-2}$) | 8.33 (+3.7%) | 12.40 (+5.7%) | 10.11 (+5.1%) | 8.08 (+4.0%) |

* All calculation are restricted to surface sites with more than 5 years observational records and drought frequency (SPEI<-1.3) greater than 10%.

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**Table S3: Fire occurrence frequency (%) and fire emissions of OC, BC, SO$_2$ and PM$_{2.5}$ under drought and normal conditions. Data are from the GFED inventory.**

| GFED (g/m² month⁻¹) | West | | | Great Plains | | | Southeast | | | Northeast | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | Dry | Nor[b] | Diff[c] | Dry | Nor | Diff | Dry | Nor | Diff | Dry | Nor | Diff |
| Fire freq[a] (%) | 41.20 | 33.00 | 1.25 | 43.40 | 35.10 | 1.24 | 86.30 | 83.90 | 1.03 | 28.20 | 23.30 | 1.21 |
| OC | 102.03 | 42.03 | 2.43 | 31.83 | 9.41 | 3.38 | 220.87 | 125.07 | 1.77 | 60.40 | 76.89 | 0.79 |
| BC | 9.55 | 3.68 | 2.59 | 3.94 | 1.07 | 3.68 | 14.09 | 9.49 | 1.48 | 3.69 | 4.56 | 0.81 |
| SO$_2$ | 14.98 | 5.99 | 2.50 | 5.58 | 1.53 | 3.65 | 26.49 | 15.86 | 1.67 | 7.12 | 8.98 | 0.79 |
| PM$_{25}$ | 201.55 | 79.12 | 2.55 | 81.99 | 21.64 | 3.79 | 321.65 | 199.88 | 1.61 | 85.51 | 106.99 | 0.80 |

[a] Fire frequency is calculated by weighting the months impacted by fire emissions with the total months.
[b] Nor refer to normal conditions.
[c] Difference is in times.

**Table S4: BC and OA concentrations (μg m⁻³) with and without fire impacts under drought and normal conditions.**

| | West | | Great Plains | | Southeast | | Northeast | |
|---|---|---|---|---|---|---|---|---|
| | BC | OA | BC | OA | BC | OA | BC | OA |
| Normal (all conditions) | 0.19 | 1.72 | 0.13 | 1.25 | 0.50 | 3.69 | 0.41 | 2.30 |
| Drought (all conditions) | 0.26 | 2.66 | 0.15 | 1.70 | 0.54 | 4.52 | 0.45 | 2.53 |
| Diff (%) | 37.64 | 54.04 | 19.64 | 36.37 | 8.79 | 22.58 | 12.19 | 10.24 |
| Normal (w/ fire) | 0.29 | 2.68 | 0.14 | 1.42 | 0.49 | 3.72 | 0.72 | 3.53 |
| Drought (w/ fire) | 0.36 | 3.91 | 0.17 | 1.98 | 0.54 | 4.59 | 0.75 | 3.55 |
| Diff (%) | 25.27 | 45.50 | 16.62 | 39.46 | 9.78 | 23.37 | 4.30 | 0.37 |
| Normal (w/o fire) | 0.14 | 1.25 | 0.12 | 1.15 | 0.51 | 3.53 | 0.31 | 1.92 |
| Drought (w/o fire) | 0.19 | 1.78 | 0.14 | 1.49 | 0.52 | 4.10 | 0.34 | 2.13 |
| Diff (%) | 35.47 | 42.33 | 18.18 | 28.86 | 3.13 | 16.13 | 10.87 | 10.98 |
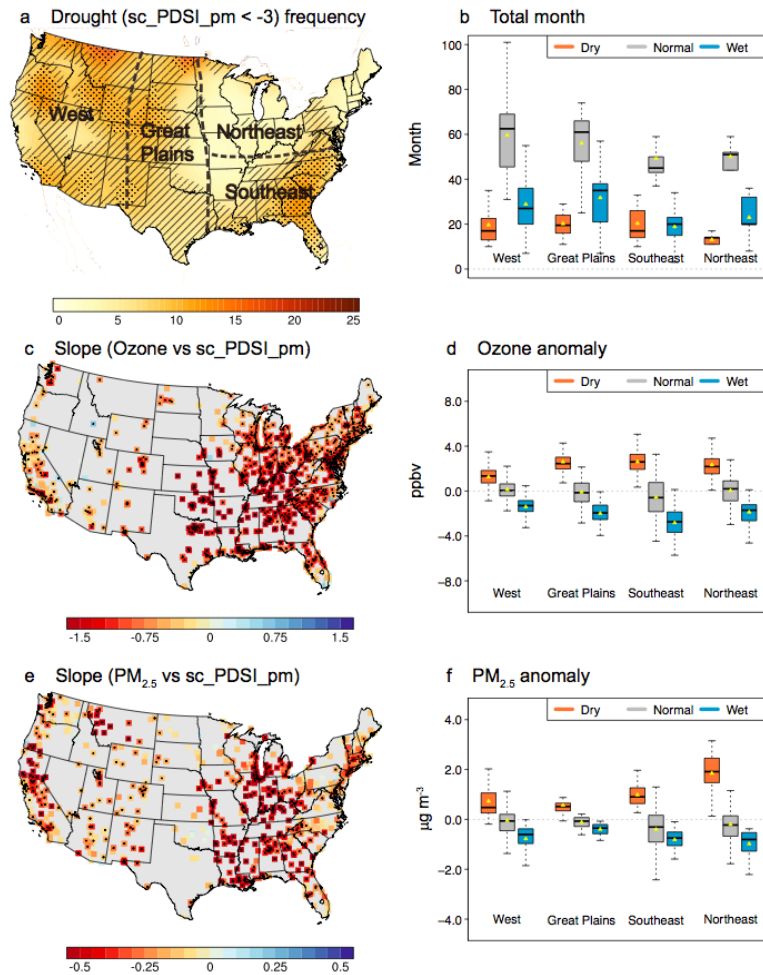
**Figure S1:** Percentage occurrence of severe drought months (sc_PDSI_pm < -3) over the continental US (a) during 1990-2014 (black dots indicate drought frequency greater than 10%, italic lines indicate drought frequency greater than 5%, and dashed lines show the division of four geographical regions). Linear regression slope of sc_PDSI_pm with $O_3$ (c) and $PM_{2.5}$ (e) anomalies at surface sites; black dots indicate regression significance at 95% level. Boxplot comparisons of the number of months (b), ozone (d) and $PM_{2.5}$ (f) at drought (sc_PDSI_pm < -3), normal (-1 < sc_PDSI_pm < 1) and wet conditions (sc_PDSI_pm > 3) by region. The triangles in the boxplot indicate mean value. All the surface data shown are restricted to sites with data records longer than 5 years and more than 5% occurrence of drought.
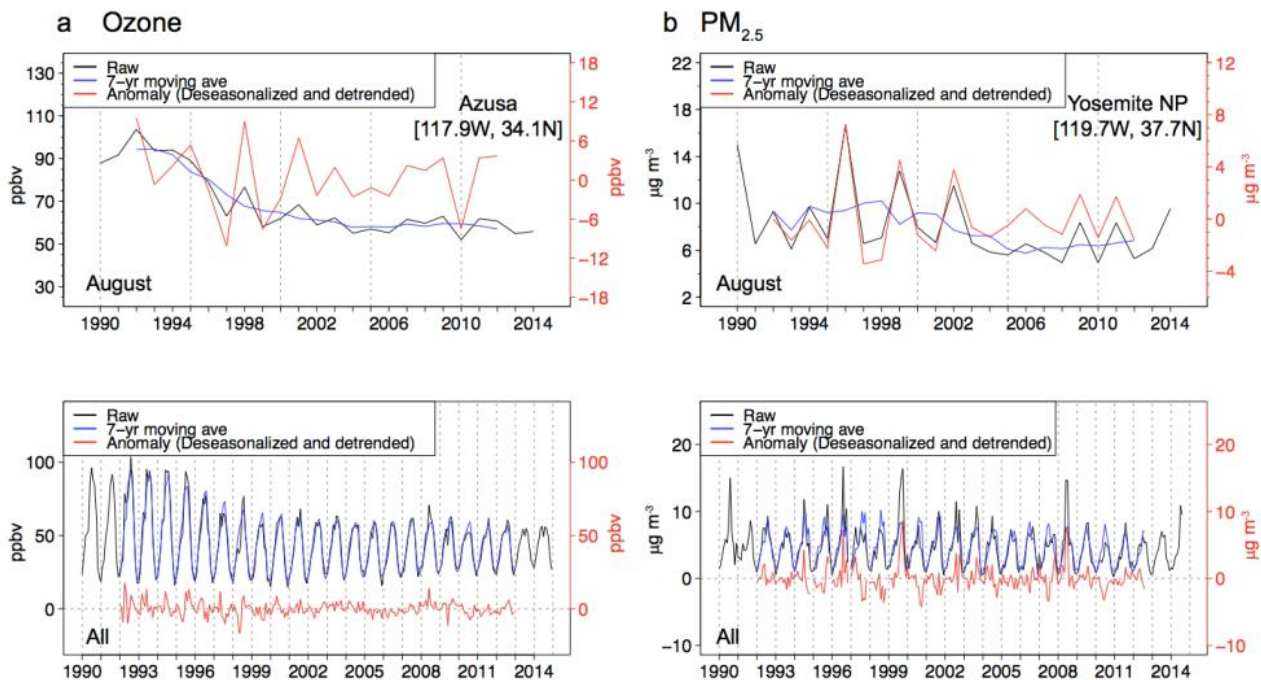
**Figure S2:** Comparison of the monthly raw data and anomalies of ozone time series at Azusa (a) and $PM_{2.5}$ time series at Yosemite NP (b). The upper panels show the data for the month of August and the lower panels for all the months during the time period of 1990-2014. The anomalies are derived by subtracting a 7-year moving average from the raw data at each month.
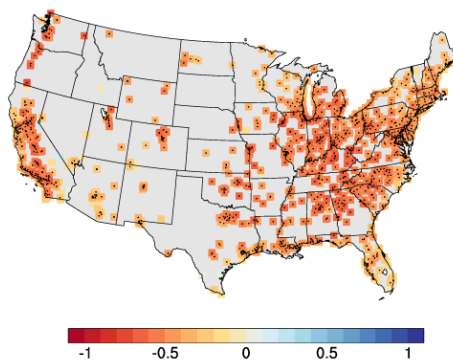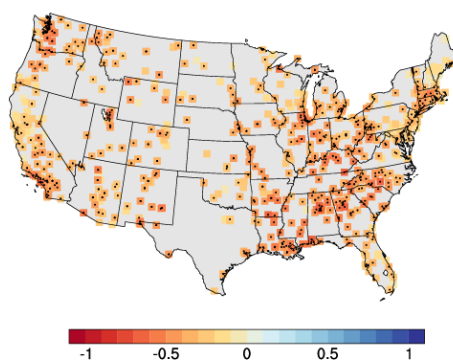
**Figure S3:** Correlation coefficient (r) of SPEI with $O_3$ (a) and $PM_{2.5}$ (b) anomalies at surface sites; black dots indicate regression significance at 95% level.
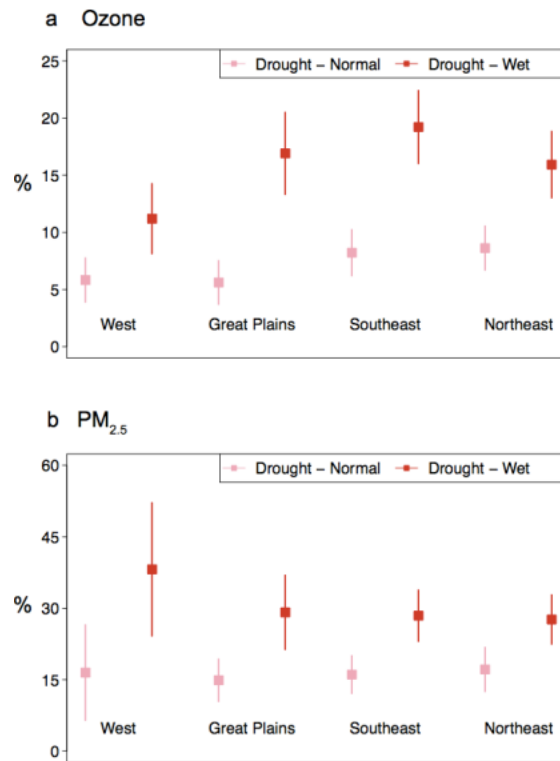
**Figure S4:** Percentage differences of ozone (a) and PM$_{2.5}$ (b) under drought (SPEI < -1.3) as compared to normal (-0.5 < SPEI < 0.5) and wet conditions (SPEI > 1.3) over the Western, Great Plains, Southeastern and Northeastern US. The error bar indicate 1/2 standard deviation. All surface sites are restricted with data records longer than 5 years and more than 10% occurrence of drought.
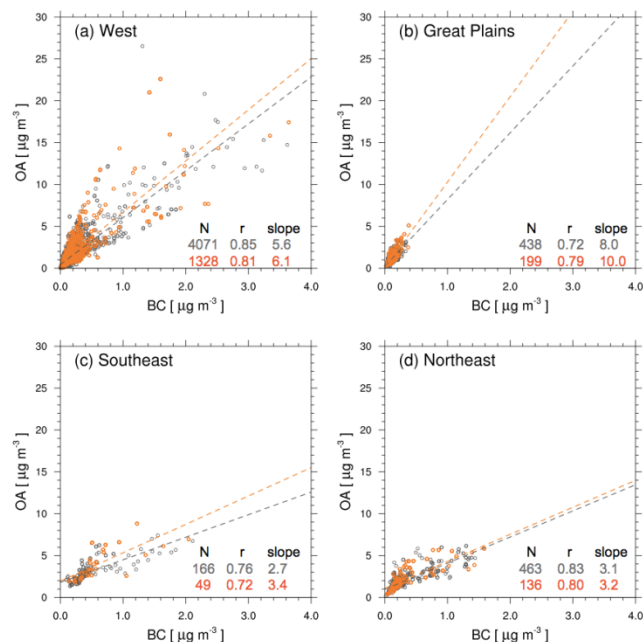
**Figure S5:** Comparison of BC and OA correlation between drought (SPEI < -1.3, color in orange) and normal (-0.5 < SPEI < 0.5, color in gray) condition without impacts from fire emissions over the Western (a), the Great Plains (b), Southeastern (c) and Northeastern (d) US. Shown in insert are the total number of data (N), correlation coefficient (r) and slope from linear regressions.
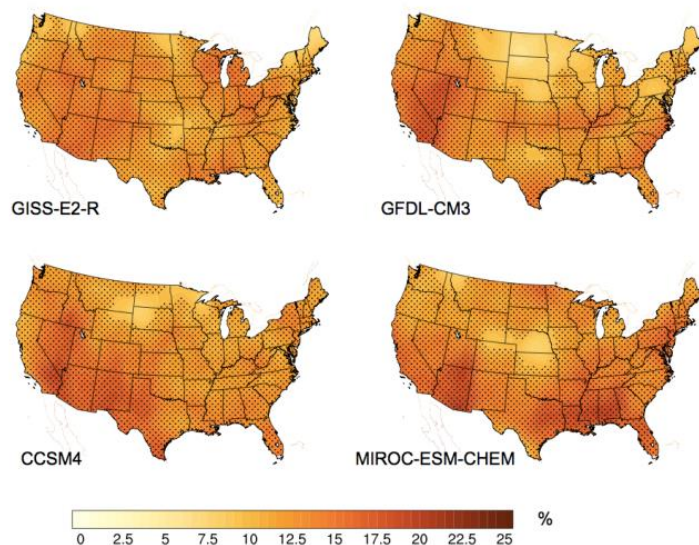
**Figure S6:** Simulated drought (SPEI < -1.3) frequency (%) by four CMIP5 models during 1990-2014 periods (data for 1990-2005 are from historical runs and data for 2006-2014 are from RCP2.6 runs).
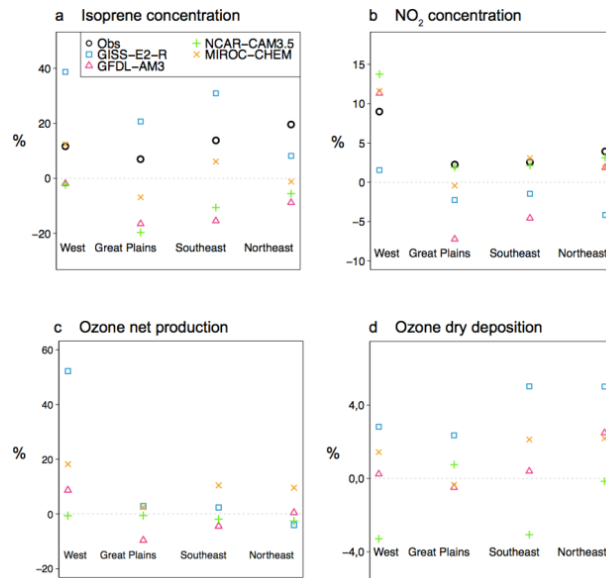
**Figure S7:** Percentage changes of surface concentrations of (a) isoprene, and (b) $NO_2$, (c) ozone net production, and (d) ozone dry deposition between drought (SPEI < -1.3) and normal (-0.5 < SPEI < 0.5) conditions over the Western, the Great Plains, the Southeastern and Northeastern US from surface observations (black circle; when applicable) and ACCMIP models (colored symbols).
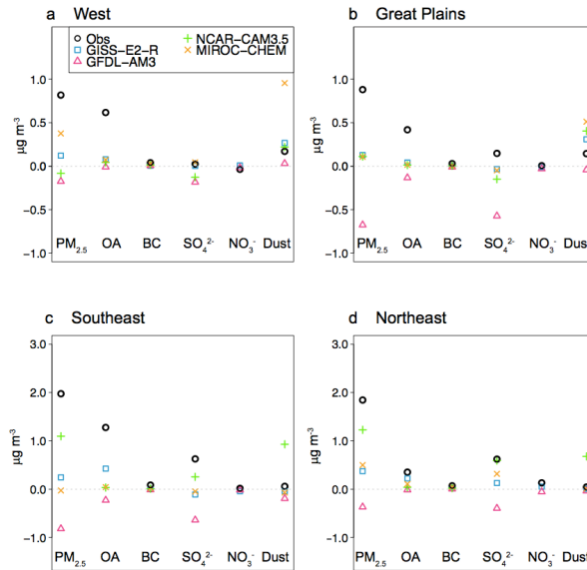
**Figure S8:** Changes of $PM_{2.5}$ speciation between drought (SPEI < -1.3) and normal (-0.5 < SPEI < 0.5) conditions over the Western, the Great Plains, the Southeastern and Northeastern US from surface observations (black circle) and ACCMIP models (colored symbols, when available). Note the changes in dust includes both coarse and fine mode.
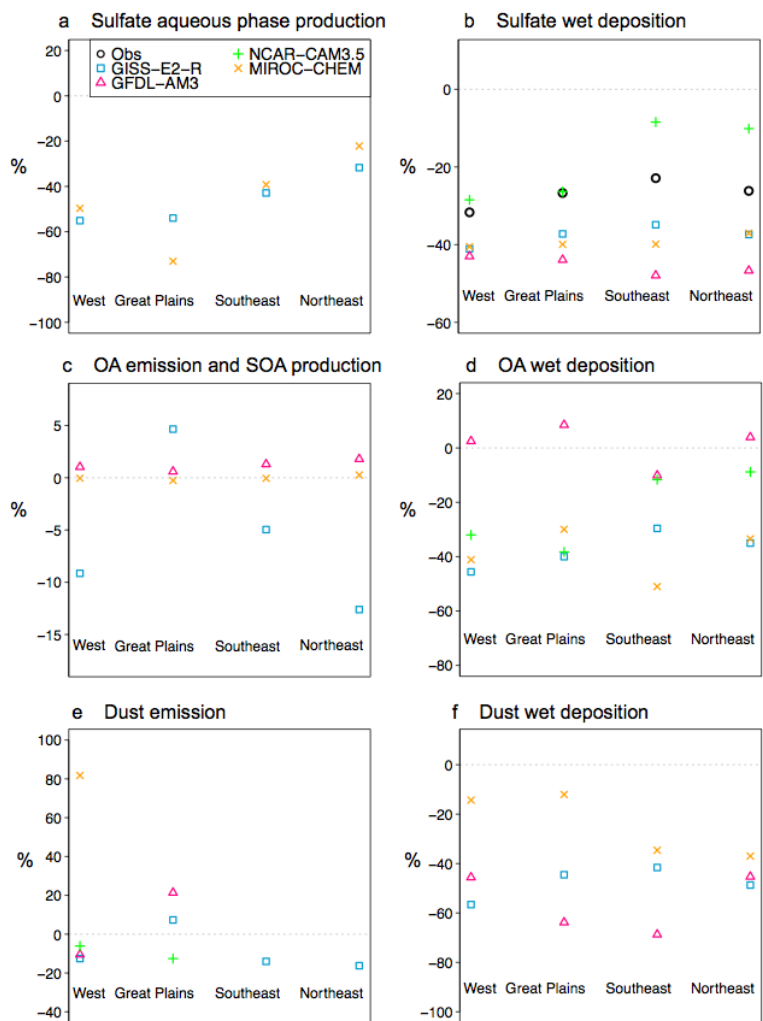
**Figure S9:** Percentage changes of sulfate production and wet deposition, OA emissions and wet deposition, and dust emission and wet deposition between drought (SPEI < -1.3) and normal (-0.5 < SPEI < 0.5) conditions over the Western, the Great Plains, the Southeastern and Northeastern US from surface observations (black circle, when available) and ACCMIP models (colored symbols, when available).