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Key Points:
- Surface O_3 was significantly enhanced in the western U.S. in June of 2015 due to multiple meteorological anomalies
- The elevated ozone caused several urban air quality sites to have many days with maximum 8 h averages over 70 ppb
- The data in June 2015 show enhanced ozone/temperature slopes at several sites, compared to previous June data

Supporting Information:
- Supporting Information S1

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Meteorological anomalies lead to elevated O_3 in the western U.S. in June 2015

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Abstract In winter through early summer of 2014–2015, temperatures in the western U.S. were substantially enhanced due to a persistent high-pressure ridge and warm water in the northeastern Pacific Ocean. Concurrently, monthly averaged maximum daily 8 h average (MDA8) O_3 in June 2015 was enhanced by 3–13 ppb across a large portion of the western U.S. At the Mount Bachelor Observatory (2.8 km above sea level) in central Oregon, O_3 in June 2015 was enhanced by 11 ppbv compared to the long-term mean. Some urban areas had many days in June 2015 with MDA8 values above the current air quality threshold of 70 ppbv. We show that the high O_3 was associated with enhanced temperatures, reduced cloud fraction, increased stagnation, and increased biogenic emissions. The data in June 2015 show enhanced $\Delta O_3/\Delta$ temperature slopes at several sites, compared to previous June data, due to these multiple factors.

1. Introduction

Ozone (O_3) is an important air pollutant that increases respiratory stress and is associated with a number of health issues, up to and including premature mortality [*Bell et al*., 2004; *Lippmann*, 1993; *Silva et al*., 2013]. In the lower atmosphere, O_3 is photochemically generated by reactions involving nitrogen oxides and hydrocarbons, which are emitted by industrial processes, combustion, and natural sources. In the U.S. nearly all regions have substantially reduced anthropogenic emissions of the precursors over the past several decades, and thus, peak O_3 concentrations have declined considerably in most areas [*Cooper et al*., 2012; *Simon et al*., 2015; *Strode et al*., 2015]. At the same time, the U.S. Environmental Protection Agency (EPA) has tightened the O_3 standard several times in response to new health data. The most recent change to the standard came in 2015, when the allowable maximum daily 8 h average (MDA8) was reduced to 70 ppb [*National Ambient Air Quality Standards for Ozone*, 2015]. The form of the standard is complex, with an area being considered in attainment of the standard if the 3 year average of the annual fourth highest MDA8 is 70 ppb or below.

Meeting the new O_3 standard will present a significant challenge for many regions of the country. One reason is that U.S. background O_3 (USB) is already a substantial fraction of the 70 ppb standard. USB is defined as O_3 formed from natural sources in the U.S. plus natural and anthropogenic sources in countries outside the U.S. [*Dolwick et al*., 2015]. Essentially, USB quantifies the natural and foreign sources of O_3 that would be difficult or impossible to control. USB varies daily and is a function of season, meteorology, and elevation. O_3 increases as a function of altitude, so higher-elevation sites are exposed to higher concentrations of O_3. For this reason, many parts of the western U.S. have seasonal average USB concentrations that are up to 70% of the 70 ppb standard [*Zhang et al*., 2011]. Studies have indicated that daily variations in USB are an important control on urban air quality in the western U.S. [*Langford et al*., 2015; *Parrish et al*., 2010; *Wigder et al*., 2013]. At the same time, USB appears to be increasing in the western U.S. [*Cooper et al*., 2010; *Gratz et al*., 2015].

Increasing temperatures, due to climate change, will also present a challenge for meeting the new O_3 standard. This is because O_3 production increases with temperature [*Sillman and Samson*, 1995] and because temperature covaries with other factors that are conducive to O_3 production and accumulation (e.g., emissions, stagnation, and solar insolation). Thus, most analyses of air quality in urban areas find that temperature is one of the most important variables to explain variations in daily O_3 concentrations [*Camalier et al*., 2007]. Many studies have attempted to predict future O_3 and health impacts in the U.S. based on estimates of future emissions, meteorology, and USB [e.g., *Bell et al*., 2007; *Chen et al*., 2009; *Avise et al*., 2012; *Fang et al*., 2013; *Fann et al*., 2015; *Pfister et al*., 2014; *Rieder et al*., 2015; *Weaver et al*., 2009]. A wide range of results have been reported, depending on the assumptions made. For example, *Chen et al*. [2009] projected an annual average increase in MDA8 O_3 of 9.6 ppb in 2050, averaged across the U.S., due largely to increases in USB using the

Intergovernmental Panel on Climate Change (IPCC) A2 emission scenario. *Pfister et al*. [2014] also projected a significant increase in U.S. surface O_3 in summer, based on future temperatures and the A2 Representative Concentration Pathway 8.5 emission scenario. In contrast, *Wu et al*. [2008] used IPCC emission scenario A1B and projected significant reductions in summertime MDA8 O_3 across the U.S., but this projected reduction is partially offset by climate change. *Steiner et al*. [2010] suggested that changes in the relationship between temperature and O_3 at very high temperatures may moderate future changes in O_3 due to climate change. *Weaver et al*. [2009] compared results from 12 global- and regional-scale models used to project future O_3 in the U.S. and found significant differences between models. Thus, model projections of future O_3 are highly uncertain and depend critically on the assumptions made with respect to U.S. and global emissions and future meteorological conditions.

While global average temperature continues to increase, this trend is not spatially or temporally uniform. Starting in late 2013, a transition from the negative to positive phase of the Pacific Decadal Oscillation brought with it exceptionally warm sea surface temperatures (SSTs) in the North Pacific [*Blunden and Arndt*, 2016; *Bond et al*., 2015; *Hartmann*, 2015; *Mote et al*., 2016]. These conditions persisted through 2014 and into 2015. In the North Pacific, the SST anomaly was 2.5 standard deviations above the long-term mean for most of 2015 [*Blunden and Arndt*, 2016]. The warm waters in the North Pacific had significant ecological impacts on marine mammals and caused extensive growth of harmful algae on the entire west coast of the U.S. in the summer of 2015 [*Blunden and Arndt*, 2016; *Cavole et al*., 2016; *McCabe et al*., 2016]. The anomalous SSTs also impacted air temperatures across the western U.S. [*Bond et al*., 2015]. *Mote et al*. [2016] argue that the 2014–2015 temperature anomaly was, in part, due to anthropogenic influence on climate change. While the anomalously high temperatures and O_3 were both present for much of 2014 and the first half of 2015, our analysis focuses on June, when USB concentrations are already high [*Gratz et al*., 2015]. For June 2015, the monthly mean temperatures across the western U.S. were enhanced by 1–6°C over the long-term mean and the monthly average MDA8 O_3 values were enhanced up to 11 ppb. During this period, many individual air monitoring stations had numerous days with MDA8 values greater than 70 ppb. Our analysis focuses on the role of temperature, cloud cover, stagnation, and biogenic emissions in explaining the high O_3 days. Based on our analysis, we conclude that O_3 was not enhanced due to an increase in USB but was associated with enhanced O_3 production from domestic emissions, combined with exceptional meteorological conditions. We also examine the slope of the O_3-temperature relationship for this unusually warm period.

2. Methods

The Mount Bachelor Observatory (MBO) is a mountain-top observatory at 2.8 km above sea level operated by the University of Washington since 2004 [*Jaffe et al*., 2005]. Ozone and other chemical tracers are measured by using standard UV methods, and calibrations are referenced to a Washington State transfer standard, which is calibrated against the EPA Region 9 Standard Reference Photometer. At the MBO a diurnal cycle in O_3, water vapor, and other tracers indicates a clear diurnal pattern in free tropospheric (FT) air versus boundary layer influence at the site. We have used water vapor mixing ratios as a means to separate FT air at the site [*Ambrose et al*., 2011]. By comparing the MBO data to radiosonde data from two nearby sites, we identify a monthly water vapor criteria. Water vapor mixing ratios below this value correspond to the FT data (Table S1 in the supporting information). Details on O_3 and other measurements at the MBO are in previously published papers [*Ambrose et al*., 2011; *Gratz et al*., 2015]. O_3 and meteorological data from other locations are taken from the EPA Air Quality System (AQS) database (https://www.epa.gov/air-data).

Satellite-derived cloud fraction are from the Atmospheric Infrared Sounder onboard NASA's Aqua satellite. Data are accessed at http://giovanni.gsfc.nasa.gov/giovanni/. For large-scale meteorological analyses, we use results from the National Centers for Environmental Prediction's North American Regional Reanalysis (NCEP NARR). This is a combined high-resolution model/data assimilation analysis of meteorological patterns covering 1979 to present. We access the results via the NOAA Earth System Research Laboratory (http://www.esrl.noaa.gov/psd/data/gridded/data.narr.html). Vertical profiles of winds were obtained from twice daily radiosonde observations at 0 and 12 Greenwich mean time. Data were accessed via the University of Wyoming atmospheric data site (http://weather.uwyo.edu/upperair/sounding.html). Backward air mass trajectories were calculated by using the NOAA Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model (http://ready.arl.noaa.gov/HYSPLIT.php) with the Global Data Assimilation System (GDAS)

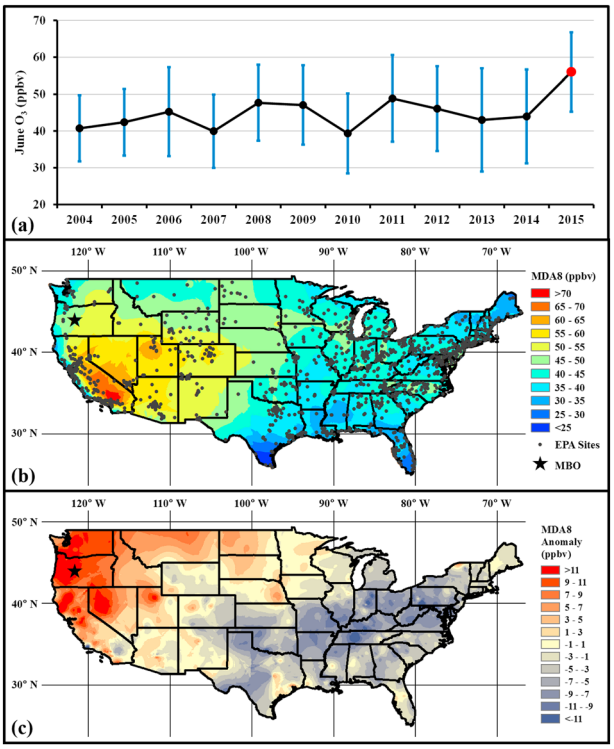

Figure 1. Data showing large changes in O_3 for June 2015. (a) Average of all O_3 data from MBO for June for 2004–2015 (the vertical bars show 1σ of the hourly values), (b) June 2015 mean MDA8 from EPA AQS stations, and (c) June 2015 MDA8 anomaly relative to previous 5 years. In Figures 1b a star is used to show location of the MBO and EPA AQS sites are shown with dots.

1° × 1° meteorological data set [*Stein et al.*, 2015]. We used a trajectory starting height of 500 m above ground level (agl) for each site, except for MBO, where a starting height of 1500 m agl was used. This reflects the fact that model terrain in mountainous regions does not reflect true topography. We have previously shown that 1500 m agl is the best height to use for MBO with the 1° × 1° GDAS data set [*Gratz et al.*, 2015]. The formaldehyde column data at a 0.25° × 0.25° grid resolution was derived from Ozone Monitoring Instrument (OMI)/AURA observations by using differential optical absorption spectroscopy technique combined with radiative transfer calculations. We access the database via Royal Belgian Institute for Space Aeronomy (http://h2co.aeronomie.be). For all spatial plots shown in this manuscript, we used Kriging methods to interpolate the data.

3. Results and Discussion

Figure 1a shows the June monthly mean O_3 (±1σ) at the MBO for 2004–2015. The June 2015 mean was 56.0 ppb, 12 ppb greater than the average of the previous 11 years. A *t* test demonstrates that the difference between the 2004–2014 mean and 2015 is highly significant ($P < 0.001$). The anomalous O_3 at the MBO was seen in both free tropospheric air and boundary layer influenced air (Figure S1 in the supporting information). In addition, other tracers, such as carbon monoxide and water vapor, were not significantly changed in 2015 (Figure S2). This indicates that neither enhanced long-range transport of pollution [*Jaffe et al.*, 2004] nor upper troposphere/lower stratospheric air mass intrusion could explain the enhanced O_3 in 2015. Figure 1b shows the June 2015 monthly averaged MDA8 from all EPA sites in the continental U.S. This shows the highest MDA8 values in Southern California extending to the Rocky Mountain region and the Pacific Northwest. Figure 1c shows the anomaly in monthly averaged MDA8 compared to previous June data (2010–2014). Because peak O_3 concentrations are declining in most urban areas of the U.S. [*Simon et al.*, 2015], we chose to compare the 2015 data with the most recent 5 years, rather than a longer time period. Here the data show that the highest anomalies in MDA8 extend from central California and across most of the western U.S. In a number of urban areas there were many days in June 2015 with MDA8 values greater than 70 ppb. For example, Salt Lake City, UT, had 16 days in June 2015 alone with MDA8 values above 70 ppb, whereas in the previous 5 years, they averaged only 2 days per month in June with values over 70 ppb.

Figure 2a shows the anomalous pattern of temperature in June 2015 at 850 mbar (approximately 1.5 km above sea level). The monthly mean temperature anomaly was more than 5°C in some regions. The pattern at the surface is nearly identical. The region with a temperature anomaly of more than 2°C extended from approximately San Francisco, CA, to the Canadian border and from central Wyoming and Utah westward to the Pacific Coast. The temperature anomaly over North America is clearly linked to the warm SSTs over the North Pacific [*Bond et al.*, 2015], but it is also linked with other factors including a high-pressure ridge, reduced winds, and decreased cloud cover, as described below. Figures S3a and S3b show the climatological June mean and June 2015 anomaly for the 850 mbar geopotential height. The figures show that the mean

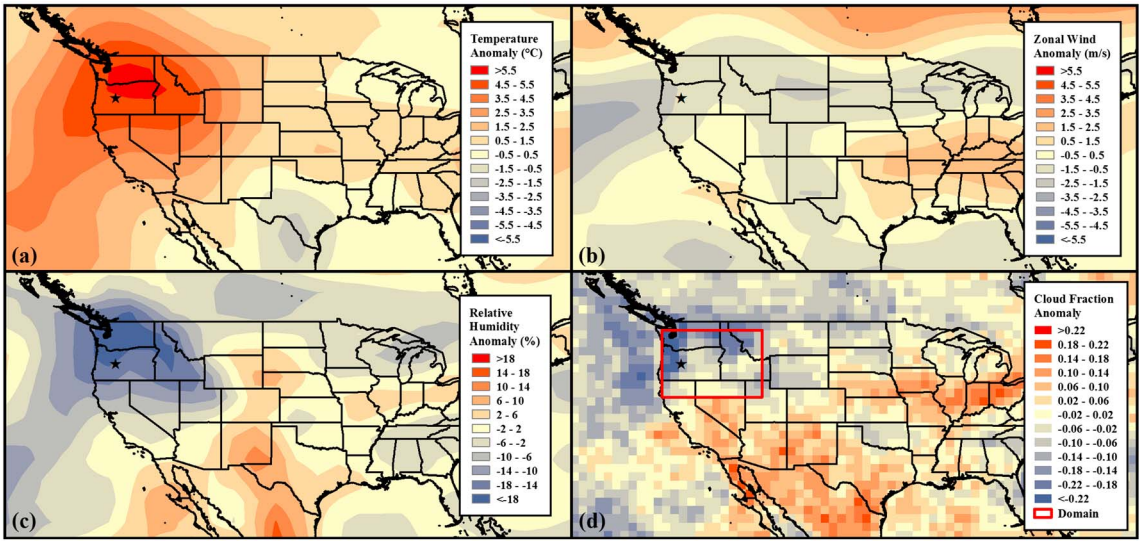

Figure 2. June 2015 meteorological anomalies: (a) North American Regional Reanalysis (NARR) 850 mbar temperature anomaly, (b) NARR 850 mbar zonal wind anomaly, (c) NARR 850 mbar RH anomaly, and (d) AIRS daytime cloud fraction anomaly.

position of the Pacific high in June 2015 has shifted to the northeast, the Aleutian low-pressure system has receded to the north, and the zone of low pressure is much weaker than usual. One important effect of this change was to shift the region of strong zonal flow to the north. Figure 2b shows the 850 mbar zonal wind anomaly for June 2015. What is usually a strong zonal or westerly wind flow into the northern United States has been reduced considerably in June 2015.

A reduction in mean wind speeds in the northwestern U.S. was confirmed by using two approaches. First, we examined vertical wind data from eight routine radiosonde sites in the northwest region that were most impacted by the change in zonal flow. The data (shown in Table S2) show that wind speeds at 0–4 km above sea level in June 2015 are, on average, 18% lower than in the previous 10 years. Second, we examined backward trajectories from the HYSPLIT model calculated for nine sites in the region. Table S3 shows these results. The trajectories also confirmed reduced air mass transport (e.g., lower average wind speeds) in June 2015. On average, the 24 h transport distances in June 2015 were reduced by 27% across all sites compared to the previous 7 years. The combination of reduced wind speeds and reduced trajectory transport distances indicates greater stagnation and accumulation of pollutants in June 2015 compared to previous years.

The enhanced temperature resulted in another important factor: reduced cloud cover. Figure 2c shows that the mean relative humidity (RH) (%) for June 2015 at 850 mbar was reduced by more than 15% across the northwest region. At the MBO, average RH was reduced by 19.7% in June 2015, compared to the previous 10 years. This reduced RH resulted in much lower cloud cover, as seen by the AIRS instrument on the NASA Aqua satellite (Figure 2d). For the Pacific Northwest region, bounded by 124–112°W and 40–48°N (box in Figure 2d), the average daytime cloud fraction in June 2015 was 0.31, compared to 0.41 in the 12 previous years. The reduction in cloud cover likely increased photochemical production of O_3 from regional sources.

We also considered two other factors that might have influenced O_3 during this time: boundary layer heights (BLH) and increased biogenic emissions in the region. Both might increase during periods of warmer temperatures. For BLH, we examined results from the Weather Research and Forecasting model, run at the University of Washington, which simulates meteorology at a resolution of 4 km. The results for June 2015 showed no consistent pattern across the model domain compared to earlier years (see Table S4). This is consistent with the results reported by *Jacob and Winner* [2009], who find no clear relationship with BLH and surface O_3.

If O_3 production is limited by volatile organic compounds (VOCs), then changes in biogenic emissions could also contribute to enhanced O_3. *Steiner et al.* [2006] used the CMAQ model and demonstrated significant O_3 sensitivity to biogenic VOCs for central California. In the Pacific Northwest, O_3 sensitivity to VOCs is usually

observed during peak O_3 periods [*Xie et al.*, 2011]. Given the strong positive temperature dependence of these emissions [*Guenther et al.*, 2012], it is possible that changes in biogenic VOCs contributed to the enhanced O_3 seen in June 2015. The enhancement in biogenic VOC emissions can be approximated by the relationships described in *Guenther et al.* [2012]. For a 4°C enhancement in temperature (see Figure 2), we would expect an increase in isoprene, sesquiterpenes, and monoterpenes of 60, 97, and 49%, respectively. To examine possible changes in biogenic emissions in 2015, we used satellite-derived formaldehyde, since this is indicative of isoprene, as well as α and β-pinene emissions, which are also abundant in the western U.S. [*Guenther et al.*, 2000; *Palmer et al.*, 2006]. Figure S4a shows the mean formaldehyde column for June 2010–2014, and Figure S4b shows the June 2015 anomaly as a % of the earlier years. While we see some evidence for enhanced formaldehyde, we note that the formaldehyde columns over the western U.S. are very low. Over the region bounded by 124–112°W and 40–48°N, column formaldehyde in June 2015 is enhanced by 39%, relative to the previous 5 years. However, caution should be used in interpreting this value as the cloud fraction in 2015 was also significantly reduced. To our knowledge, there are no other isoprene, terpene, or formaldehyde data for the region for this time period. In summary, biogenic VOCs are likely important as O_3 precursors in the western U.S., and we see some evidence for enhanced biogenic emissions based on the OMI formaldehyde columns in June 2015.

The results above suggest that the enhanced O_3 seen in June 2015 was likely due to enhanced local and/or regional photochemical production, rather than enhanced transport of anthropogenic precursors from regional, global, or background sources. This is confirmed by examining tracers of pollution and upper troposphere/lower stratospheric (UTLS) sources of O_3, as measured at the MBO. Based on past analysis of MBO data [*Ambrose et al.*, 2011], carbon monoxide (CO), aerosols, and water vapor mixing ratios are good tracers for global pollution or UTLS sources. For June 2015, the average CO, aerosol scattering coefficient, and water vapor mixing ratios were 111.4 ppb, 7.4 Mm^{-1}, and 4.5 g/kg, compared to long-term means for June of 110.5 ppb, 4.2 Mm^{-1}, and 4.4 g/kg. So while the average June 2015 temperature at the MBO was enhanced by 5.4°C and the % relative humidity decreased by 19.7%, the CO and water vapor mixing ratios were essentially identical to their long-term means. Aerosols are likely enhanced due to the lower frequency of clouds observed in 2015. At the MBO, we saw no evidence for enhanced pollution (e.g., enhanced CO or aerosols) or UTLS (decreased water vapor mixing ratio) in June 2015 (Figure S2). We also evaluated water vapor mixing ratios by using the same eight radiosonde sites, as described earlier. For 0–4 km above ground level, the June 2015 water vapor mixing ratios were slightly greater (6.8%) compared to the long-term mean. These data are shown in Table S5. Thus, both the MBO data and the radiosonde data from eight sites in the western U.S. indicate that the enhanced O_3 in June 2015 was likely of domestic origin and not due to global background sources, such as long-range transport of pollutants or UTLS intrusions.

Figure 3 shows the relationship between MDA8 O_3 and the daily maximum temperature for four representative sites in the western U.S. for June data in 2010–2015, with data for 2015 shown in red. We use data from only the past 5 years to minimize effects from changing emissions. While all of the data demonstrate a relationship between MDA8 O_3 and the daily maximum temperature, the relationship in 2015 is rather different. The 2015 data show the enhanced temperatures; however, O_3 values are also higher at the same temperature compared to previous years. The $\Delta O_3/\Delta$temperature linear regression slopes are greater in 2015 for all sites. The difference is statistically significant for two of the four sites ($P < 0.05$, Sacramento and Seattle) (see Table S6). At 35°C the O_3-temperature relationships suggest an additional 4–18 ppb of O_3 at these four sites.

Our conclusions differ somewhat from those reported by *Steiner et al.* [2010], who suggested that climate change impacts on O_3 may be ameliorated by a reduced $\Delta O_3/\Delta$temperature slope at the highest temperatures (daily maximum temperature greater than 39°C). For the sites considered in Figure 3, only Sacramento shows any days with daily maximum temperatures at this level or above. Despite the fact that the average daily maximum temperature in Sacramento in June 2015 was 3.1°C higher, compared to previous years, there were only 4 days with a maximum temperature of 39°C or greater. So while the results of *Steiner et al.* [2010] show a reduction in the $\Delta O_3/\Delta$temperature slope above 39°C, we see few days with such a high daily maximum temperature and no reduction in the slope in June 2015. Likely, the 2015 data show the combined effects of temperature, winds, and solar insolation, and so any small reductions in O_3, due to high temperatures, are offset by the impact of reduced wind speeds and solar insolation. The data in Figure 3

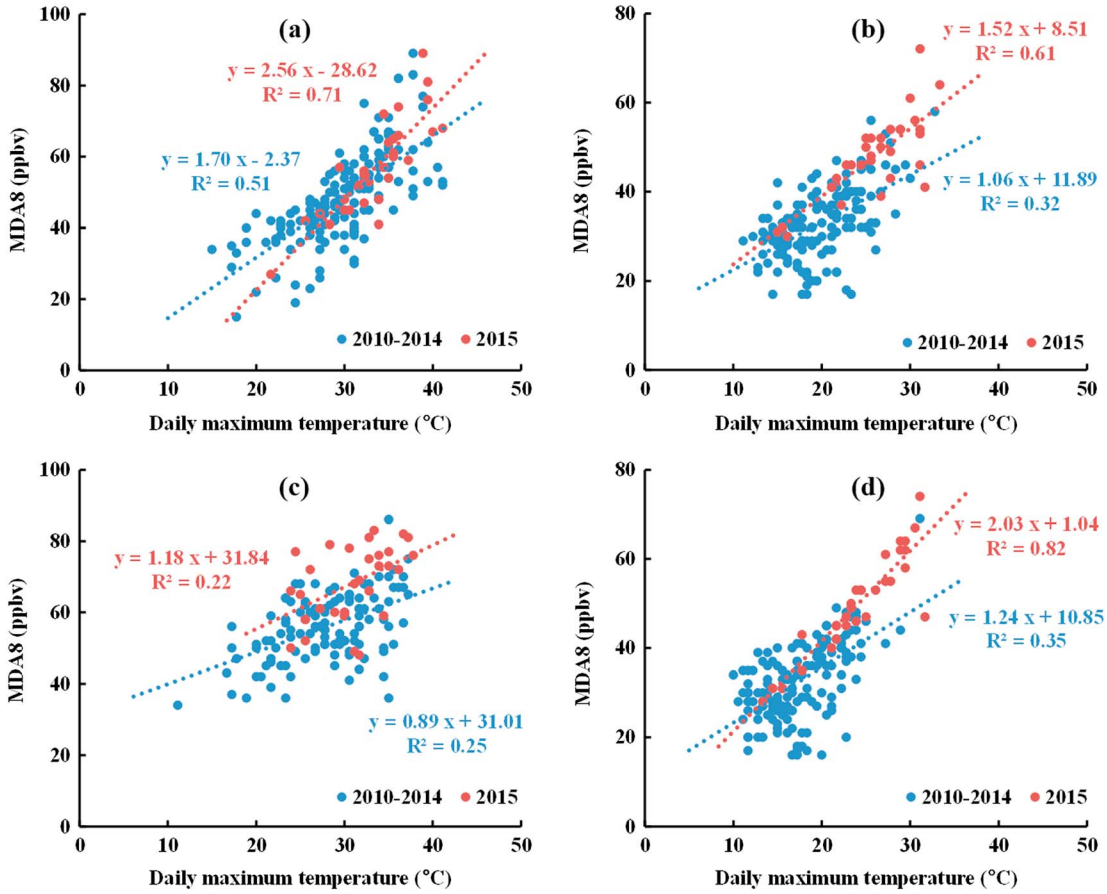

Figure 3. MDA8 versus daily maximum temperature for individual days in June 2010–2014 (blue) and June 2015 (red) for (a) Sacramento, CA (AQS id: 06-067-0006); (b) Portland, OR (AQS id: 41-005-0004); (c) Salt Lake City, UT (AQS id: 49-035-3006); and (d) Seattle, WA (AQS id: 53-033-0023).

indicate that temperature alone did not cause the enhanced O_3, but that reduced wind speeds and cloud cover, plus possibly enhanced biogenic emissions, also contributed.

5. Conclusions

Surface O_3 in the U.S. may increase or decrease in the future. Continued emission reductions in the U.S. will tend to reduce concentrations, but these may be offset by rising temperatures and/or increasing international emissions. In addition, understanding the interannual variations in O_3 is important to fully comprehend the implications of the 3 year average in compliance metrics (i.e., the 3 year average of the fourth highest MDA8), since a single high year can put a region out of compliance with the standard. We have shown how changes in temperature, wind speeds, and cloud cover combined to cause an unusually high O_3 year in the western U.S. During June 2015 alone, some urban sites in the western U.S. had as many as 16 days above the 70 ppb threshold. Our analysis indicates that this enhanced O_3 in June 2015 was due to O_3 production from domestic sources caused by the anomalous meteorological conditions. Understanding how much of the O_3 change was due to each factor individually will require detailed atmospheric modeling. At the same time, a comparison of chemical models with the 2015 O_3 season would help improve our predictions of future changes in O_3.

Acknowledgments
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