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Characterizing changes in surface ozone levels in metropolitan and rural areas in the United States for 1980–2008 and 1994–2008

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ABSTRACT

In this analysis, we characterize urban and rural ozone (O_3) trends across the US for the periods 1980–2008 (29 years) and 1994–2008 (15 years) using three exposure metrics, which summarize daily O_3 concentrations to reflect different ways O_3 may affect human health and vegetation. We observe that a statistically significant trend at a specific monitoring site, using one exposure metric, does not necessarily result in a similar trend using the other two metrics. The two most common trends among the monitoring sites are either a continuation of negative trending over the 29-year period or a shift from negative to no trend status, indicating a leveling off of the trending. Very few sites exhibit statistically significant increases in the exposure indices. In characterizing the statistically significant changes in the distribution of hourly average O_3, we observe subtle statistically significant changes in the lower part of the distribution (i.e., below 50 ppb) that are not necessarily captured by the trending patterns associated with the three exposure metrics. Using multisite data from 12 metropolitan cities, we find that as the frequency of higher hourly average concentrations is reduced, the lower hourly average concentrations also move upward toward the mid-level values. The change in the number of the hourly average concentrations in the lower range is consistent with decreased NO scavenging. We recommend assessing possible subtle shifts in O_3 concentrations by characterizing changes in the distribution of hourly average concentrations by month. Identifying statistically significant monthly changes in the mid- and low-level hourly average concentrations may provide important information for assessing changes in physical processes associated with global climate change, long-range transport, and the efficacy of models used for emission and risk reductions. Our results indicate that it is important to investigate the change in the trending pattern with time (e.g., moving 15-year trending) in order to assess how year-to-year variability may influence the trend calculation.

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1. Introduction

The US EPA has reported that emission reduction in ozone (O_3) precursors has been substantial over the past 29 years (US EPA, 2009a). For the period 1980–2008, the percent change in emissions for nitrogen oxides (NO_x) and volatile organic compounds (VOC) was 40% and 47%, respectively. Due to these emission reductions, many of the higher hourly average O_3 concentrations experienced in the 1980s have been reduced (US EPA, 2006). Although progress has been made in reducing the peak hourly average O_3 concentrations at many nonattainment areas in the US, several areas have not met deadlines and have been reclassified to higher nonattainment categories (e.g., Atlanta, GA; Baton Rouge, LA; Beaumont-Port Arthur, TX; Ventura Co, CA; and Houston-Galveston-Brazoria, TX). In addition, the US EPA has recently proposed additional areas be changed to higher O_3 reclassification categories for failing to meet the mandated attainment deadlines (US EPA, 2009b).

Lefohn et al. (2008) investigated the relative reduction trend in the peak and mid-level (50–99 ppb) hourly average O_3 values by applying health- and vegetation-based exposure metrics for the periods 1980–2005 and 1990-2005. The two human health-based metrics used by Lefohn et al. (2008) focused on high concentrations in the hourly average O_3 distribution, while the vegetation metric (W126) included a weighted cumulative average across the entire distribution. This 24-h seasonal W126 exposure metric, described in Lefohn and Runeckles (1987) and Lefohn et al. (1988), uses a sigmoidal weighting to emphasize the more relevant higher

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concentrations. Using these three exposure metrics for assessing trending, Lefohn et al. (2008) reported that most of the sites across the US exhibited decreasing or no trending in the exposure indices investigated. Few sites exhibited increasing (i.e., positive) trending patterns. For sites with declining O_3 levels, a pattern of rapid decreases in the higher hourly average concentrations, but much slower decreases in mid-level concentrations, was observed.

Besides applying the three exposure metrics, Lefohn et al. (2008) also investigated the statistically significant monthly trends by characterizing the temporal changes in the distribution of the hourly average O_3 concentrations. By this method, it may be possible to identify physical and chemical processes that dominate particular portions of the distribution during a particular time of the year. For example, Oltmans et al. (2006) reported that the continuous monitoring site at the 3400 m altitude Mauna Loa Observatory (MLO) in September and October exhibited changes in the distribution of hourly values with occurrences in the 20–30 ppb range declining while hourly values in the 40–50 and 50–60 ppb increased. The authors reported that it was likely that the change in the autumn months was associated with a shift in transport characteristics of air masses reaching the island of Hawaii. In addition, quantification of changes in the hourly distribution patterns and emission reductions may inform judgments concerning control program efficacy and health or environmental risk. At some sites, Lefohn et al. (2008) noted that, while the three exposure metrics exhibited no general trending, there were monthly distributional changes.

Using data for the periods 1980–2008 (29 years) and 1994–2008 (15 years), this analysis explores whether historical trending patterns at specific sites are continuing or changing. We focus in greater detail on characterizing trending patterns for sites located in 12 major metropolitan cities and in 15 rural areas. In addition, we explore the consistency of trending at specific O_3 monitoring sites by characterizing patterns over moving 15-year periods.

2. Approach

Extreme value hourly, 8 hourly, and cumulative 24 hourly exposure metrics were used in this analysis: (1) an annual 2nd highest daily maximum 1-h average concentration, (2) an annual 4th highest daily maximum 8-h average concentration, and (3) a cumulative 24-h average sigmoidally weighted W126 concentration. Study data were downloaded from the US EPA's Air Quality System (AQS) database for the period 1980–2008. For a specific year to be included in the analysis, at least 75% of the hourly average O_3 values within the year and the O_3 monitoring season (US EPA, 2009c) had to be valid. For higher latitude sites in the US an O_3 monitoring season is less than twelve months; in Colorado, the O_3 season is 7 months (March–September), while in southern California the O_3 season for many of the sites is 12 months. A series of additional data validation steps was followed for specific metrics. Data validation for the hourly metric required that (1) at least 18 of 24 hourly values had to be valid to compute a valid daily maximum and (2) at least 75% of the possible daily maximum 1-h averages for each year within an O_3 season had to be valid to compute a valid 2nd highest daily maximum. Data validation for the 8-h metric required that (1) at least 6 of 8 hourly values had to be valid across 8 contiguous hours to compute a valid daily 8-h maximum, (2) at least 18 of 24 daily 8-h averages had to be valid within a calendar day to compute a valid daily maximum, and (3) at least 75% of the possible daily maximums for each year within an O_3 season had to be valid to compute a valid 4th highest daily maximum. The time series for trend calculations had to satisfy the following two further data capture criteria: (1) at least 22 (75%) valid years for 1980–2008 and 12 (75%) valid years for 1994–2008 periods and (2) beginning with the last five years of record (i.e., 2004–2008), at least three of five valid years were required within each 5-year block of years.

The W126 metric was also adjusted for missing values as follows: (1) the monthly value of each metric was calculated if at least 75% of the hourly data were available for a specific month (a corrected monthly cumulative metric was calculated as the uncorrected monthly cumulative metric divided by the fractional data capture), and (2) if a month with less than 75% data capture had the two adjacent months each having at least 75% data capture (a corrected monthly cumulative metric with less than 75% data capture was calculated as the arithmetic mean of the corrected monthly cumulative metrics for the two adjacent months). A year with more than one such interpolation was not considered valid. If all of the months contained within an O_3 season had valid estimates (by the method described above) of the corrected monthly cumulative metric, the corrected seasonal cumulative metric was calculated as the sum of the corrected monthly cumulative metrics. Otherwise, a valid estimate of the corrected seasonal cumulative metric was not reported. The criteria for trend calculation for a time series of values were the same as for the hourly and 8-h health metrics.

Following trend characterization for those sites that met the data capture criteria for the 1980–2008 and 1994–2008 periods, we compared *by site* the trending patterns for the 29-year period (1980–2008) and 15-year period (1994–2008). In addition, we also assessed the consistency of trends at specific monitoring sites over moving 15-year periods (e.g., 1986–2000, 1987–2001, 1988–2002, 1989–2003, 1990–2004, 1991–2005, 1992–2006, 1993–2007, and 1994–2008).

In its assessment of the 8-h O_3 standard, the US EPA (2007) evaluated 12 cities in its human health risk analysis (i.e., Atlanta, Boston, Chicago, Cleveland, DC, Detroit, Houston, Los Angeles, New York, Philadelphia, Sacramento, and St. Louis). We identified specific monitoring sites located in these 12 cities over the 1994–2008 period which had sufficient data capture. The initial process was to identify candidate monitoring sites in these cities by using the O_3 design value tables published by the US EPA on its web site (US EPA, 2009d) for the period 2006–2008. A design value is a statistic that describes the air quality status of a given area relative to the level of the National Ambient Air Quality Standards (NAAQS). The design value is the 3-year average of the annual 4th highest daily maximum 8-h O_3 concentration at a given site. Following identification of the candidate sites, we confirmed that they had sufficient data capture for the 1994–2008 period and characterized their trends. Investigators have characterized the changes in O_3 levels at rural monitoring sites for assessing possible impacts from long-range transport that may be associated with Asian emissions on surface O_3 concentrations in the US. It is assumed that although some of these rural sites are affected by local and regional anthropogenic emissions, they are useful for assessing changes associated with transport from Asia. In addition, rural monitoring sites are useful for assessing changes in O_3 precursor emission reductions. In our analysis, we have characterized the trending at 15 rural O_3 monitoring sites.

Because the assessed sites do not necessarily experience a statistically significant trend for each month in the O_3 season, we also characterized the monthly trends. The monthly trends were characterized as changes in frequency within each 10 ppb increment (i.e., bin) of the distribution; the 10 ppb bin data were arranged as a time series by month using the same valid data criteria as for the metrics described previously.

The Theil estimate (Hollander and Wolfe, 1999) was used to estimate the trend slope for all cases noted above. The Theil estimate is a non-parametric estimator that is numerically identical to the ordinary least squares (OLS) slope estimate when the OLS model assumptions are satisfied. The Theil estimate is determined as the median of slope estimates calculated as the slope of the line passing through paired points for all point pairs in the data set of interest. To test for statistical significance, Kendall's tau test (Lefohn and Shadwick, 1991; Hollander and Wolfe, 1999) was used to determine significance at the 10% level. Because the tail probability can change abruptly from year to year, the significance level of 0.10 was selected to reflect the degree of variability for the Kendall's τ statistic over the range of years in the time series.

3. Results

3.1. General observations across the US using the three exposure metrics

Tables 1 and 2 summarize the number of valid site metric trends characterized as negative (declining) trend, no trend, or positive (increasing) trend for the periods 1980–2008 and 1994–2008. For 1980–2008, there were more sites exhibiting decreasing trends than no trend for the 1-h, 8-h, and W126 metrics. For the 1-h and 8-h metrics for this period, there was a tendency for the largest %/year decreases to occur in southern California (Figs. 1a and 2a), while for the W126 index, the largest %/year decreases occurred in southern California, the Midwest, the Northeast, and southern US (Fig. 3a). The figures provide quantitative information about the trending patterns for specific monitoring sites. There were few sites with statistically significant increases observed for the 1980–2008 period (Table 1).

For the 1-h metric, there were more numbers of negative trend monitoring sites for the 1994–2008 period than no trend sites. For the same period for the 8-h metric, there were approximately equal numbers of sites experiencing trends and no trends. For the W126 metric, there were about half as many negative (35%) as no trend (62%) sites. For all three metrics, there were very few sites exhibiting statistically significant increases (Table 2). For the 1-h metric, there was a tendency for the largest %/year decreases to occur in southern California, the Midwest, the Northeast, and southern US (Fig. 1b). For the 8-h index, there was a tendency for the largest %/year decreases to occur in southern California, the Midwest, and the Northeast (Fig. 2b). For the W126, the largest %/year decreases occurred in southern California, the Midwest, and the eastern US (Fig. 3b). Similar to the 1980–2008 trending results, the figures provide quantitative information about the trending patterns for specific monitoring sites.

In order to identify whether O_3 trending patterns changed over the 29-year period, we compared the 1980–2008 with the 1994–2008 trending patterns on a *site-by-site* basis for those sites that met the data capture criterion for both periods. This contrasts with the results of Tables 1 and 2 and Figs. 1–3, where we included all sites that met the data capture criterion for either 1980–2008 or 1994–2008. The predominant patterns were a (1) negative trend to no trend status, (2) no change in negative trend status, and (3) no change in no trend status (Table 3). For many monitoring sites when comparing 1980–2008 to 1994–2008, a shift from negative to no trend status was observed. This reflects a decrease in the higher hourly average concentrations in the early years followed by much slower decreases in the later years. In reviewing results for all three exposure metrics, we found that approximately half the monitoring sites shifted from negative to no trend status.

Table 1
Summary of the number of monitoring sites that experienced negative (decreasing), no, or positive (increasing) trends for the 2nd highest 1-h average, 4th highest 8-h average, and the 24-h W126 cumulative exposure index for 1980–2008.

Trending direction	1980–2008		
	Exposure metrics		
	1-h	8-h	W126
Negative Trend	218 (87%)	177 (71%)	105 (56%)
No Trend	32 (12%)	66 (27%)	81 (42%)
Positive Trend	1 (1%)	5 (2%)	3 (2%)
Total	251	248	189

Table 2
Summary of the number of monitoring sites that experienced negative (decreasing), no, or positive (increasing) trends for the 2nd highest 1-h average, 4th highest 8-h average, and the 24-h W126 cumulative exposure metric for 1994-2008.

Trending direction	1994–2008		
	Exposure metrics		
	1-h	8-h	W126
Negative trend	376 (58%)	329 (51%)	191 (35%)
No trend	268 (41%)	308 (48%)	341 (62%)
Positive trend	2 (1%)	5 (1%)	18 (3%)
Total	646	642	550

3.2. Trend patterns observed in 12 metropolitan cities for the 1994–2008 period

A summary for the 15-year period, 1994–2008, of the trending patterns observed for the 12 metropolitan cities (i.e., Atlanta, Boston, Chicago, Cleveland, DC, Detroit, Houston, Los Angeles, New York, Philadelphia, Sacramento, and St. Louis) is provided in Table 4 and Table S1 (supplementary material). The information in Table 4 is provided for illustrative purposes and complete details are provided in Table S1. In the tables, the 1-h, 8-h, and seasonal W126 O_3 trends are quantified by %/year (Theil estimate). The months that experienced statistically significant trends in the changes in the distribution of hourly average concentrations and the 8-h design values (DV) are also presented in the tables. Where the change in concentrations in a specific monthly bin moved higher, the month notes a "+" sign with a "−" sign when the bin change moved lower. Where both the higher hourly average concentrations and the lowest concentration bin (i.e., 0–9 ppb) decreased, meaning that both the high and low concentrations moved toward the center of the distribution, the specific month notes a "±" sign.

Atlanta, Boston, Chicago, DC, Houston, Los Angeles, New York, Philadelphia, Sacramento, and St. Louis are currently US EPA O_3 nonattainment areas. Detroit and Cleveland have attained the 8-h 1997 O_3 standard. For 2006–2008, Rockdale, GA (AQS 132470001) is the 8-h design value site for Atlanta (95 ppb). Over 1994–2008 the Rockdale site did not experience a statistically significant trend for any of the three O_3 exposure metrics (Table S1). Two other sites (i.e., Fulton and Paulding) in the Atlanta area with sufficient data did exhibit 1–2 %/year decreases in the 1-h and 8-h O_3 trending metrics (Table 4). The Fulton County (GA) site has shifts in hourly average O_3 concentrations from higher (110–130 ppb) to mid-level bins (40–70 ppb) (Fig. 4). Shifts from the 0–20 ppb range to the mid-level bins were observed; this shift is consistent with reduced NO scavenging associated with NO_x emission reductions.

In most cases, O_3 monitoring sites in the Boston area did not experience statistically significant changes in the three metrics for the period (Table S1). However, individual months from 1994–2008 exhibited shifts in the distributions of the hourly average concentrations. In some months, the concentration shifts were from the mid-range to the higher hourly average O_3

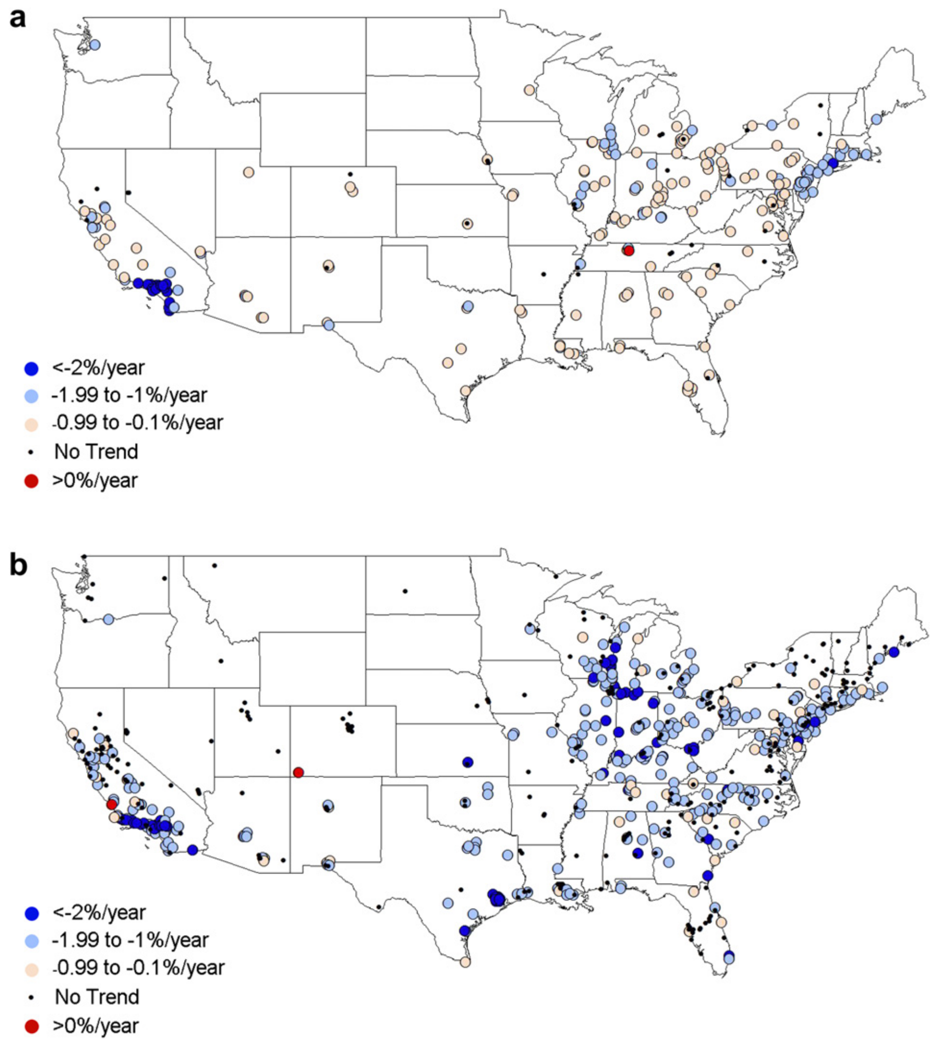

Fig. 1. Trend of 2nd highest 1-h average ozone metric for (a) 1980–2008 and (b) 1994–2008.

concentrations; in other months, the concentration shift was in the opposite direction. For Bristol County (MA), the only site that experienced a change (−1%/year) in one of the O_3 trending metrics (1-h), Fig. 5 illustrates the shifts in the hourly average concentrations. Many of the Chicago (IL) sites exhibited negative trends in the 1-h and 8-h average concentration metrics (−1 to −2%/year) (Table S1). However, no trends were observed at most sites for the 24-h seasonal W126 O_3 metric. Most of the sites experienced shifts in the distributions of the hourly average concentrations. In some cases, the hourly average concentrations in the lowest bin (i.e., 0–9 ppb) shifted into the higher-level concentration bins (i.e., 30–40 ppb). Also, there were instances when both the higher hourly average concentrations and the lowest concentrations (0–9 ppb) moved toward the mid-level values.

Although there were no sites with positive trending using the three exposure metrics, the Cleveland (OH) and DC areas did not experience consistent patterns of negative or no trending over the 15-year period (Table S1). However, almost all monitoring sites did exhibit statistically significant changes by month in the distributions of the hourly average concentrations. Monitoring sites located in Cleveland exhibited instances when both the higher hourly average concentrations and the lowest concentrations (0–9 ppb) moved toward the mid-level values. For the Detroit (MI) area, statistically significant decreases (−1 to −2%/year) for the 1-h metric occurred (Table S1). However, no change was observed for the 8-h and 24-h seasonal W126 O_3 metrics. All monitoring sites exhibited statistically significant changes by month in the distributions of the hourly average concentrations.

The Houston (TX) area exhibited consistently decreasing (−2 to −4%/year) trending of O_3 concentrations for the 1-h and 8-h concentration exposure metrics (Table S1). In many cases, the 24-h seasonal W126 O_3 metric exhibited decreasing trends (−4 to −6%/year). Statistically significant changes by month in the distributions of the hourly average concentrations were observed for all sites. For the period 1994–2008, Fig. 6 shows a site in Harris County that exhibited statistically significant changes from the high hourly average concentrations (i.e., 120–170 ppb) to the mid-range concentrations (i.e., 30–60 ppb) the month of September. More modest changes occurred during the months of April and August.

Similar to Houston (TX), Los Angeles (CA) experienced fairly consistent decreases in the 1-h and 8-h concentration metrics (−1 to −3%/year) (Table S1). About 50% of the sites experienced decreasing trends (−2 to −5%/year) in the W126 index. However, one site (AQS 060371301), located in Lynwood (CA), while exhibiting a negative trend for the 29-year period, exhibited a positive trend in the 24-h seasonal W126 exposure metric (21%/year) for the 15-year period (1994–2008). The W126 exposure index values for 1998–2002 were fairly low (i.e., below 2.5 ppm-h) in contrast to

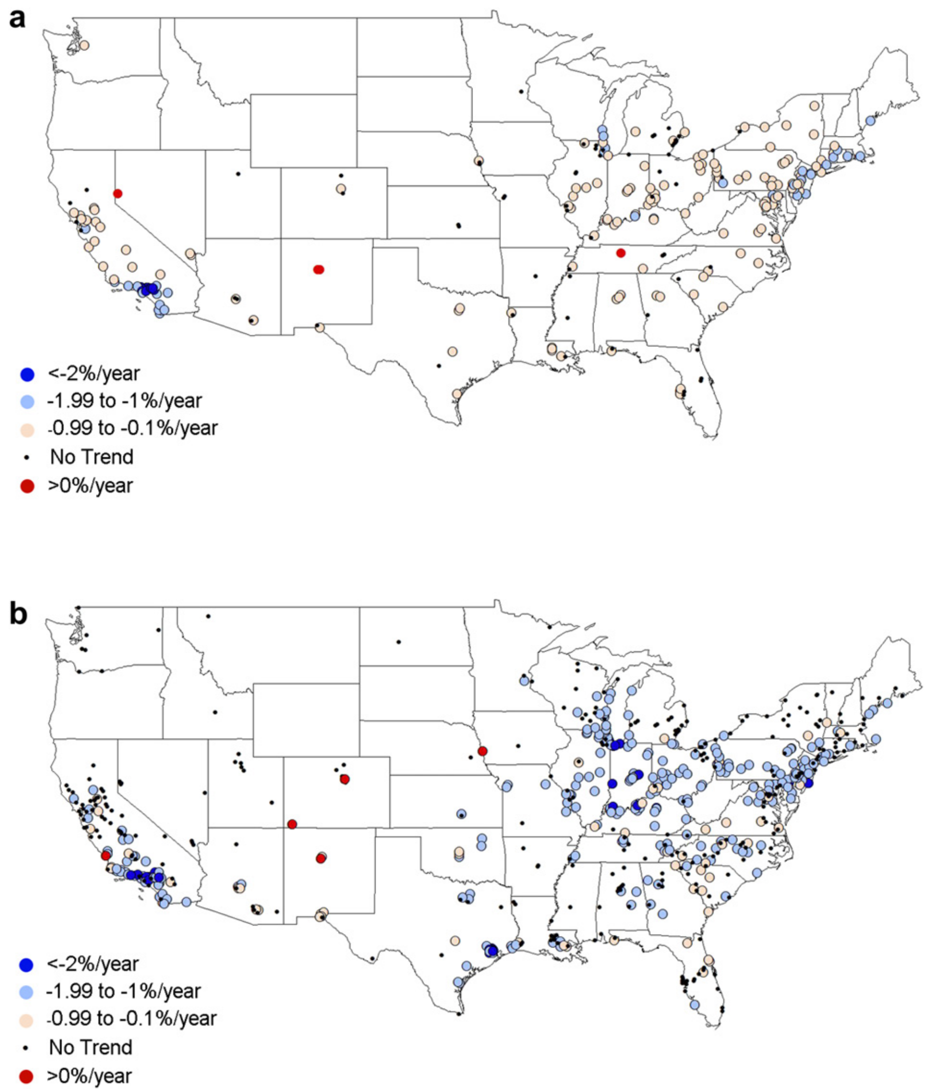

Fig. 2. Trend of 4th highest 8-h average ozone metric for (a) 1980–2008 and (b) 1994–2008.

the higher values exhibited during 2003–2006. In 2007, the W126 value (6.2 ppm-h) declined to approximately the 2003 value (5.5 ppm-h), which was much lower than the 2006 value (16.1 ppm-h). The data criteria were not met for calculating the W126 index for 2008. This site experienced a low 58 ppb design value for 2006–2008. Statistically significant positive monthly distributional changes of the hourly average concentrations were observed at this site. The southern California Lake Gregory monitoring site at Crestline (AQS060710005) usually experiences the highest O_3 design value in the US. At this site for 2006–2008, the 8-h design value was 119 ppb, the highest in the US. Over the 1980–2008 and 1994–2008 periods, this site has experienced statistically significant decreases in O_3. Fig. 7 illustrates the monthly1994–2008 statistically significant hourly distributional changes by month. Statistically significant changes from high hourly average concentrations (i.e., 100–210 ppb) to the mid-range concentrations (i.e., 50–70 ppb) occurred.

The New York City (NY) area has two sites with negative (−1 to −2%/year) trending and two sites with no trending over the 15-year period (Table S1) for the 1-h and 8-h metrics. For the W126 index, two sites exhibited negative trending (−3%/year), while the other two did not show a trend. However, almost all monitoring sites did exhibit statistically significant hourly distributional changes by month. Many sites in the Philadelphia (PA) and St. Louis (MO) areas exhibited negative (−1 to −2%/year) trends for the 1-h and 8-h exposure metrics. For the W126 index, decreases occurred in the range of −2 to −5%/year in Philadelphia and about −3%/year in the St. Louis area. In addition, all monitoring sites exhibited statistically significant monthly hourly distributional changes. Sacramento (CA) did not show statistically significant trends for the three metrics for most of the sites. However, all sites did exhibit statistically significant monthly hourly distributional changes.

3.3. Trends in rural areas

Table 5 summarizes findings of the rural O_3 monitoring sites shown in Fig. 8 for 1994–2008. Some of the monitoring sites are located in national parks (NP). The sites were selected because (1) investigators have characterized the changes in O_3 levels at rural monitoring sites for assessing possible impacts from long-range transport from Asia on surface O_3 concentrations in the US (Yienger et al., 2000; Jaffe et al., 2003; Parrish et al., 2004; Jaffe and Ray, 2007) and (2) rural monitoring sites are useful for assessing changes in O_3 precursor emission reductions. Although those

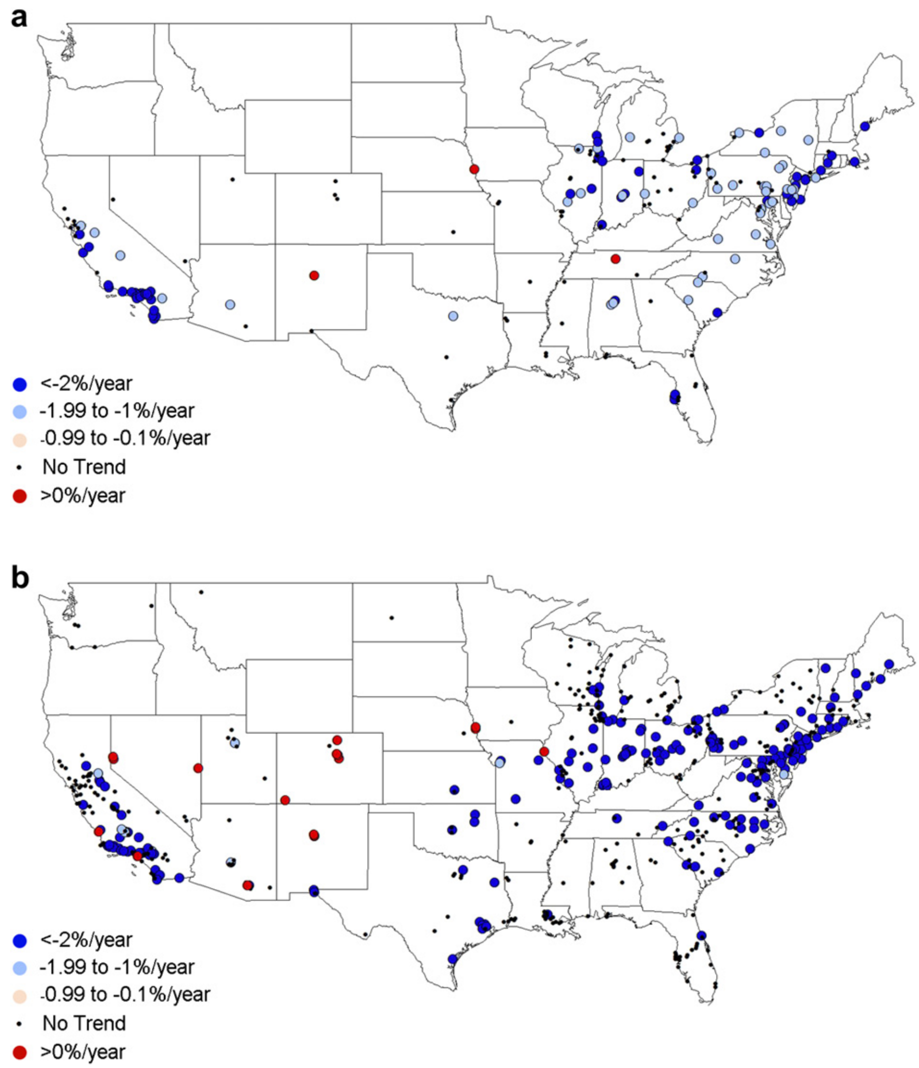

Fig. 3. Trend of 24-h W126 ozone metric for (a) 1980–2008 and (b) 1994–2008.

researchers, interested in springtime Asian transport, used 12 months of trend data, we used trend data from both urban and rural sites in the US EPA AQS database, where much of the data are not recorded for the full 12 months but for the EPA defined O_3 season (US EPA, 2009c). For the rural CA and AZ sites identified in Fig. 8 (i.e., Lassen Volcanic NP and Yosemite NP, Grand Canyon NP), the O_3 season is defined as a 12-month period. For the other rural sites identified in Fig. 8, the O_3 season is less than 12 months.

Using 12-month data from Lassen Volcanic NP, Yosemite NP, and Grand Canyon NP, we identified no exposure metric trending patterns for the 1994–2008 period (Table 5). However, for Lassen NP, for the months of January and February, there was a shift upwards from the hourly average O_3 concentrations in the 30–40 ppb range to the 50 to 60 ppb range (Fig. 9). The Yosemite NP site has a design value of 88 ppb, which violates the 1997 O_3 standard for 2006–2008. For this site for the months of January–April, there was a shift from the hourly concentrations in the 30 to 50 ppb range to the 60 to 70 ppb range (Fig. 10). For the month of May, there was a shift of the hourly values from the 30 to 50 ppb range to the 60 to 100 ppb range. In August and September, the hourly concentrations shifted downward from 100 to 110 ppb hourly values to the 60 to 70 ppb range. For Grand Canyon NP, there was an hourly upward shift during April from 50 ppb to 70 ppb.

At the Denali NP site in Alaska, there were no trending patterns observed for the 1-h and 8-h metrics but a positive trend (4%/year) for the 24-h seasonal W126 metric (Table 5). No months experienced statistically significant trending. At the Mount Rainier NP (WA) site, there were neither trending patterns observed for the three metrics (Table 5) nor statistically significant monthly changes.

Table 3
Comparison of trending patterns by site for 1980–2008 versus 1994–2008.

Comparison of trending patterns by site	Exposure metrics		
1980–2008 versus 1994–2008	1-h	8-h	W126
Negative Trend to No Trend	76 (30%)	73 (29%)	60 (32%)
Negative Trend to Negative Trend (no change)	141 (56%)	103 (41%)	44 (23%)
No Trend to No Trend (no change)	23 (9%)	41 (16%)	62 (33%)
No Trend to Negative Trend	9 (3%)	24 (10%)	13 (7%)
Positive Trend to No Trend	0	2 (1%)	0
Negative Trend to Positive Trend	1 (1%)	1 (1%)	1 (1%)
Positive Trend to Positive Trend (no change)	0	1 (1%)	1 (1%)
Positive Trend to Negative Trend	1 (1%)	2 (1%)	1
No Trend to Positive Trend	0	1 (1%)	5 (3%)
	251	248	187

Table 4
Characterization of trend patterns for selected metropolitan cities by site for 1994–2008.

		%/Year Trend				
County	AQS ID	1-h	8-h	W126	Months[a] with Trend	DV[b]
Atlanta						
Fulton	131210055	−1.45	−1.82	NS	3 (+), 4 (+), 7(−), 8 (±), 9 (+)	91
Paulding	132230003	−1.85	−1.28	–	3 (+), 7 (−), 8 (−), 9 (−)	80
Boston						
Bristol	250051002	−1.05	NS	NS	4 (+), 6 (+), 8 (−)	80
Chicago						
Cook	170310032	−2.32	−1.46	NS	4 (+), 5 (+), 7 (+), 10 (+)	74
Cook	170310072	−2.36	−2.22	−3.70	4 (+), 6 (±), 7 (±), 8 (±), 10 (±)	67
Cleveland						
Medina	391030003	−1.67	−1.35	−4.27	6 (−), 8 (−)	72
DC						
DC	110010025	−1.12	−1.26	−3.11	4 (+), 6 (+), 7 (−), 8 (+), 9 (+), 10 (+)	80
Detroit						
Wayne	261630019	−1.44	NS	NS	4 (+), 5 (+), 6 (+), 7 (+), 8 (+), 9 (+)	82
Houston						
Harris	482010051	−3.03	−2.36	−5.43	4 (−), 8 (±), 9 (−)	80
Los Angeles						
Los Angeles	060371301	NS	NS	+21.38	1–9 (+), 11 (+), 12 (+)	58
San Bernardino	060710005	−1.98	−1.84	−1.89	1 (+), 2 (+), 4 (±), 6 (±), 7 (−), 8 (−), 9 (−)	119
New York						
Suffolk	361030002	−1.22	−1.19	−3.38	6 (+), 7 (−), 8 (±), 9 (+), 10 (+)	85
Philadelphia						
Montgomery	420910013	−1.80	−1.14	−2.80	4 (+), 6 (−), 7 (±), 8 (±), 10 (+)	84
Sacramento						
Placer	060610006	−0.84	NS	NS	4 (±), 5 − 6 (+), 7 (±), 8 − 10 (+), 12 (+)	90
St. Louis						
St. Charles	291831004	−1.33	−1.02	−2.89	7 (−), 10 (−)	82

NS, Not significant.
(+) indicates a change in concentrations from a lower to a higher bin.
(−) indicates a change in concentrations from a higher to a lower bin.
(±) Both the higher hourly average concentrations and the lowest concentration bin (0–9 ppb) decreased, meaning that both the peak and the low concentrations moved toward the center of the distribution.
[a] Months with Trend, for example Jan, Feb, March,…December are designated as 1, 2, 3,…12.
[b] DV = Design Value in units of ppb for 2006-2008 as calculated by US EPA (2009d).

For the Canyonlands NP (UT) site, no trends were observed for the three metrics (Table 5) but there were subtle upward hourly June shifts from the 40–50 ppb to the 60 ppb concentration bins. A trend in the distribution was observed in October for the Craters of the Moon National Monument (ID) site (Table 5). The hourly average concentrations shifted downward from 50 ppb to 30 ppb. For the Rocky Mountain NP (CO) site, no trends were observed for the three metrics, although there were April–June upward hourly shifts. In April there was a shift from 40–50 ppb to 60–70 ppb ranges; in May from 50 ppb to 60 ppb; in June from the 30 ppb range toward the 60 ppb ranges. For the Mesa Verde NP (CO) site, positive trends were observed for the 1-h (0.73%/year), 8-h (0.75%/year) and W126 (5%/year) metrics (Table 5) and consistent hourly shifts for April, May, June, and September (Fig. 11). The shifts occurred from the 30 to 50 ppb ranges upward to the 60 to 80 ppb ranges.

For the four Great Smoky Mountains monitoring sites in North Carolina and Tennessee, there was a negative trending in the 1-h and 8-h metrics (−1 to −1.5%/year). One site experienced a W126 trending decrease (−3.5%/year). For all sites, the months with statistically significant shifts in the hourly average O_3 concentrations exhibited downward changes. For the Whiteface Mountain (NY) site, no trends were observed for the 1-h and 8-h metrics (Table 5). No months exhibited statistically significant hourly distributional changes. For the Shenandoah NP (VA) site, negative trends were observed for the 1-h (−1%/year), 8-h (−1%/year), and W126 (−4%/year) metrics (Table 5). June and July shifts occurred from the 70 to 80 ppb range downward to the 50 to 60 ppb range.

4. Discussion

In our trends analysis, we quantified the (1) trending patterns across the US for the periods 1980–2008 and 1994–2008, using 3 exposure metrics and (2) changes in distribution patterns by month for specific monitoring sites. In this section, it is our intent to place into perspective the observed trending changes and how they may be of interest to those who wish to link these changes to changes in physical processes (e.g., Oltmans et al., 2006) that may be associated with global climate change or long-range transport, as well as changes associated with O_3 precursor emission reductions. The identification of O_3 monitoring sites that exhibit changes in concentrations, such as those sites described in our analysis, provide important information to those interested about the effects of anthropogenic and natural perturbations on surface O_3 concentrations.

Many of the predictions associated with global change have indicated that surface O_3 concentrations are expected to increase in future years (Forster et al., 2007; US EPA, 2009e; Zeng et al., 2010). In addition, some researchers have indicated that long-range transport from Asia may increase surface O_3 concentrations in the US (Yienger et al., 2000; Jaffe et al., 2003; Parrish et al., 2004; Zhang et al., 2010).

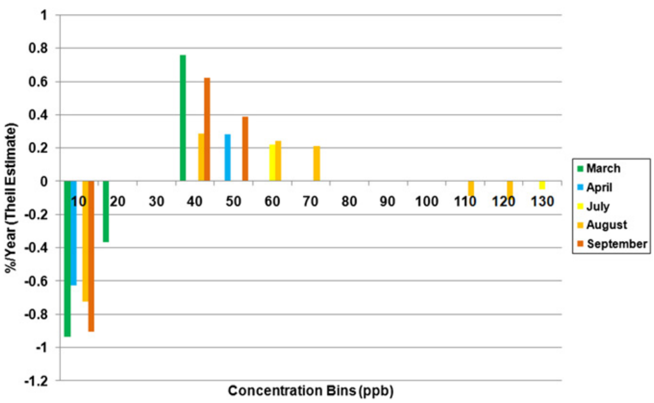

Fig. 4. Distribution of changes by month for a monitoring site located in Fulton County, GA (AQS 131210055) for 1994–2008 for the months with statistically significant changes.

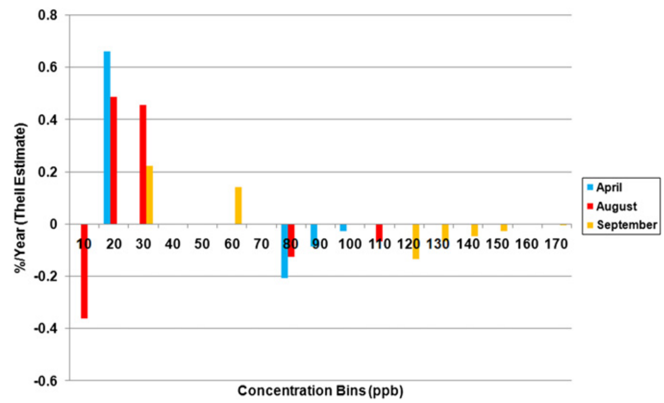

Fig. 6. Distribution of changes by month for a monitoring site located in Harris County, Texas (AQS 482010051) for 1994–2008 for the months with statistically significant changes.

In this analysis, as well as the analysis described by Lefohn et al. (2008), the application of the 1-h, 8-h, and W126 exposure metrics at times resulted in variable trending patterns (see Tables 4, 5, and S1) both temporally and spatially (see Figs. 1–3). The sites in our analysis show a consistent pattern of either decreasing or no trends. For all three metrics, there were very few sites exhibiting statistically significant increases (Tables 1–5 and S1). Thus, predictions of increasing surface O_3 concentrations that may be associated with long-range transport from Asia or global climate change were not observable using the three exposure metrics used in our analysis (Table 5). For those interested in relating the time series of emission changes with surface O_3 concentrations, when comparing changes in trending patterns on a site-by-site basis for the 1980–2008 with the 1994–2008 periods (Table 3), our results indicate that the majority of monitoring sites either (1) changed from negative to no trend, (2) remained negative, or (3) remained in the no trend status. Few sites changed from either (1) no trend to negative or (2) no trend to positive when compared over these periods. For sites mostly experiencing statistically significant declines, rapid decreases in the higher hourly values in the early years were followed by slower rates of decline. These slower rates appear responsible for a ~45% shift from negative to no trending status, depending upon the metric.

Some researchers (US EPA, 2009e; Zeng et al., 2010) have predicted that increases in surface O_3 concentrations in the northern hemisphere for specific months (e.g., January–April) may occur as a result from climate change and or stratospheric recovery. Our analysis for both urban and rural monitoring sites has focused on trending changes in the distribution of hourly average concentration by month. Figs. 4–7 provide an illustration for some of the urban sites. As the higher hourly average concentrations are reduced, hourly average concentrations in the lower end of the distribution move upward toward mid-level values. In other words, both ends of the distribution of hourly average concentrations move toward mid-level values as discussed earlier by Lefohn et al. (1998). The movement of hourly values toward the 30–60 ppb range appears to be occurring at many of the sites we investigated in this analysis. In addition, for sites exhibiting statistically significant increases during specific months (see Tables 4 and S1), the shifts occur from the lowest part of the distribution to mid-range, consistent with a reduction in NO scavenging as a result of emission reductions (US EPA, 2006). Figs. 9–11 illustrate changes in the distribution of hourly average concentrations by month for some of the rural monitoring sites. Tables 4, 5, and S1 provide information for specific sites on the trending pattern for statistically significant trends by month that occurred in the distribution of the hourly average concentrations.

In some cases, where we observed negative trending patterns for some of the metrics, we observed only *increasing* trends in the monthly distributions. For example, a monitoring site in Chicago (AQS 170310032) (see Table 4) illustrates that while negative trending occurred for the 1-h and 8-h exposure metrics, the hourly

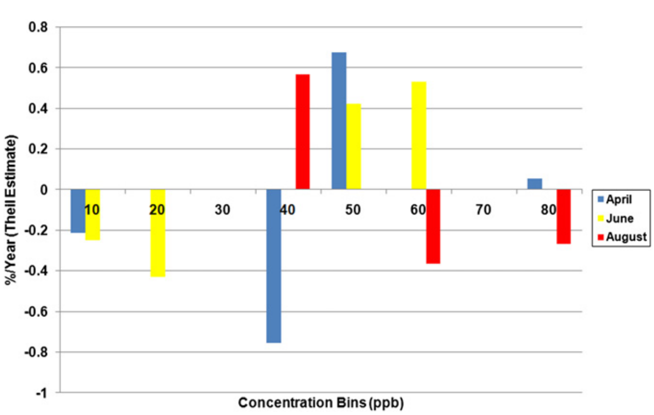

Fig. 5. Distribution of changes by month for a monitoring site located in Bristol County, MA (AQS 250010002) for 1994–2008 for the months with statistically significant changes.

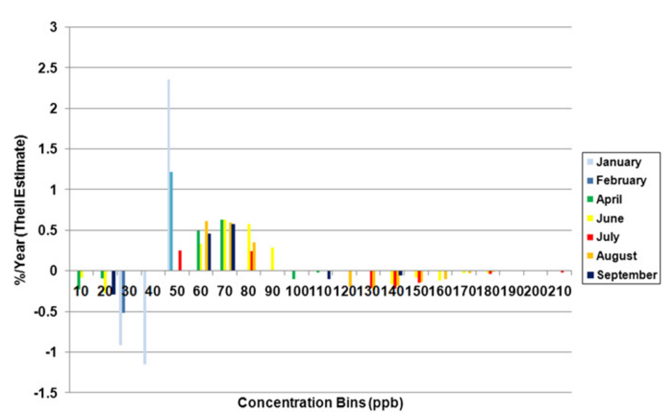

Fig. 7. Distribution of changes by month for a monitoring site located in San Bernardino County, California (AQS 060710005) for 1994–2008 for the months with statistically significant changes.

Table 5
Characterization of trend patterns for selected rural sites for 1994–2008.

Site name	AQS ID	%/Year Trend 1-h	8-h	W126	Months with Trend[a]	DV[b]
Denali NP, AK	022900003	NS	NS	+4.09		58
Mount Rainier NP, WA	530531010	NS	NS	NS		58
Lassen Volcanic NP, CA	060893003	NS	NS	–	1 (+), 2 (+)	77
Yosemite NP, CA	060430003	NS	NS	NS	1–5 (+), 8 (–), 9 (–)	88
Canyonlands NP, UT	490370101	NS	NS	NS	6 (+)	71
Craters of the Moon NM, ID	160230101	NS	NS	–	10 (–)	–
Grand Canyon NP, AZ	040058001	NS	NS	NS	4 (+)	70
Rocky Mtn NP, CO	080690007	NS	NS	NS	4 (+), 5 (+), 6 (+)	76
Mesa Verde NP, CO	080830101	+0.73	+0.74	+5.32	4 (+), 5 (+), 6 (+), 9 (+)	71
Great Smoky Mtns NP, NC	370870036	–1.53	NS	–	8 (–)	77
Great Smoky Mtns(a) NP, TN	470090101	NS	–1.26	NS	7 (–), 10 (–)	85
Great Smoky Mtns(b) NP, TN	470090102	–1.08	–0.90	–	7 (–)	72
Great Smoky Mtns(c) NP, TN	471550101	–1.49	–1.18	–3.42	7 (–), 9 (–)	82
Whiteface Mtn, NY	360310002	NS	NS	–		76
Shenandoah NP, VA	511130003	–0.98	–1.12	–3.80	6 (–), 7 (–)	75

NS, Not significant.
(+) indicates a change in concentrations from a lower to a higher bin.
(–) indicates a change in concentrations from a higher to a lower bin.
NP = National Park; NM = National Monument.
(a)Look Rock;(b)Cades Cove; (c)Cove Mountain.
[a] Months with Trend, for example Jan, Feb, March,…December are designated as 1, 2, 3,…12.
[b] DV = Design Value in units of ppb for 2006–2008 as calculated by US EPA (2009d).

average concentrations shifted from the lower values to the higher values for the months of April, May, July, and October. We believe that this may be associated with the concentration shifts from the more numerous values in the lower bins to the mid-level concentrations. We hypothesize that shifts from the higher concentrations to the mid-level values contributed to the negative trending. However, due to the smaller number of higher hourly average concentrations, we were unable to identify statistically significant decreases in this range of the distribution.

Our trending results for the O_3 monitoring site at Denali NP appear to be affected by a long-range transport event that occurred in April 2008. Although there were no trending patterns observed for the 1-h and 8-h metrics, a positive trend was observed for the 24-h seasonal W126 metric. Fig. 12 illustrates the time series over the 1994–2008 period for the 24-h seasonal W126 metric. Over the 15-year period, the highest 24-h W126 value was experienced in 2008 and this value affected the trending pattern. Oltmans et al. (in press) describe the effects of Eurasian biomass burning transport in April 2008 on surface O_3 levels in the western US. As Oltmans et al. (in press) observed, during April 2008, as part of the International Polar Year, a number of ground-based and aircraft campaigns occurred in the North American Arctic region that observed widespread gaseous and particulate biomass burning effluent. High seasonal O_3 readings were recorded at surface O_3 monitoring sites from northern Alaska to northern California. At Barrow, Alaska, the highest April O_3 readings in 36 years were recorded at the surface (i.e., hourly values > 55 ppb) on April 19, 2008. At Denali NP in central Alaska, an 8-h average of 79 ppb was recorded and elevated O_3 (i.e., >60 ppb) persisted almost continuously from April 19–23.

Investigators have reported trend results from mid-latitude sites in North America, continental Europe, and Japan, including some of the sites analyzed here (Logan et al., 1999; Ordóñez et al., 2007; Oltmans et al., 2006; Chan and Vet, 2009). Chan and Vet (2009) report trends for different regions of Canada and the United States over the 10-year time period 1997–2006 using data from

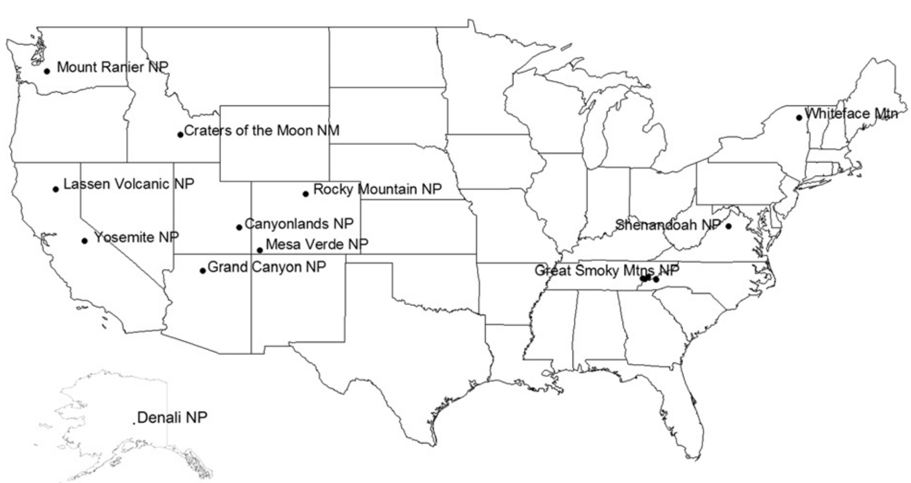

Fig. 8. Location of rural ozone monitoring sites characterized.

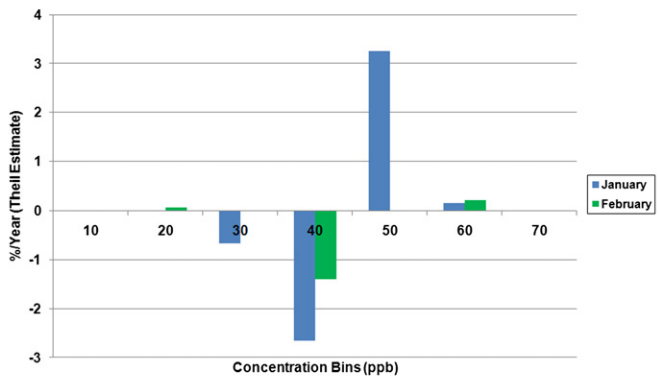

Fig. 9. Distribution of changes by month for a monitoring site located in Shasta County, California (AQS 060893003) for 1994–2008 for the months with significant changes.

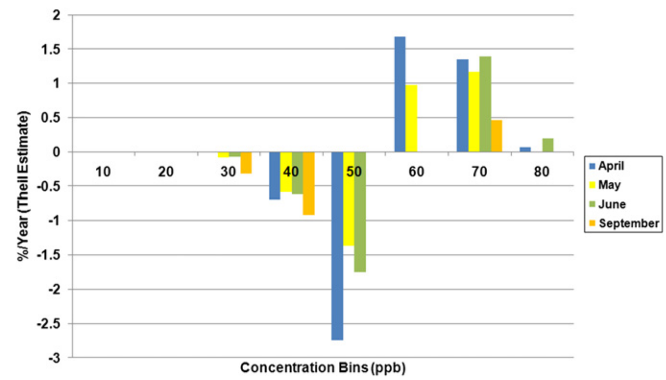

Fig. 11. Distribution of changes by month for a monitoring site located in Montezuma County, Colorado (AQS 080830101) for 1994–2008 for the months with significant changes.

non-urban monitoring sites. The authors report that O_3 trends decreased significantly in southeastern Canada and the eastern US. The authors report a statistically significant increase in southwestern British Columbia, but no significant increase in California. A comprehensive assessment of ground-level O_3 in Europe for various O_3 metrics found a strong relationship with interannual weather conditions (EEA, 2009). While modeled O_3 behavior, based on reduced emissions, matched the measured O_3 behavior for Europe as a whole, this overall picture was not replicated on a regional basis (EEA, 2009). A similar picture was found by Jonson et al. (2006), where there were important regional variations in the relationship of O_3 changes to precursor reductions over Europe. Increases in winter season O_3, primarily the mid to lower portion of the distribution, showed a stronger response to NO_x reductions than decreases in the summer (Jonson et al. (2006). A study of the influence of East Asian emissions on spring season O_3 levels at a number of Japanese sites (Tanimoto et al., 2009) found relatively small increases at lower altitude locations, but substantially larger increases at higher altitude locations between 1998 and 2007. The larger increases at higher altitudes sites were attributed to Asian emissions, however, O_3 increased more rapidly after 2003 than predicted, suggesting a significant underestimation of precursor emission increases. In Southern China, O_3 has increased in response to NO_x increases in fast developing coastal regions (Wang et al., 2009). The observed increase in O_3 levels is very likely the result of the increase in the emission of O_3 precursors in upwind source regions in the eastern coastal regions of China (Wang et al., 2009).

Locations reporting significant increases in the 1970s and 1980s appear to show leveling off or in some cases, declines in more recent decades (Oltmans et al., 2006; Derwent et al., 2007). Small O_3 increases have recently been reported at four Alpine sites for the 1995–2007 period in contrast to small reductions in precursor concentrations at these sites (Gilge et al., 2010). The O_3 increases occurred during winter months with small decreases during the summer. Therefore, because of the observed change in rate of trending, characterizing a trending pattern starting with the beginning of monitoring and ending with the last recorded may not accurately describe the observed changes in trending rates. Thus, multiple trending patterns may be characterized using shorter trending periods (e.g., moving 15-year trending periods) over the period of record. As an example of this approach, we investigate the trending for two sites (Lassen Volcanic NP and Rocky Mountain NP) using moving 15-year periods.

While no trending patterns were observed using the 1-h and 8-h extreme value metrics for Lassen Volcanic NP (CA) for the 1994–2008 period, statistically significant hourly distributional changes were observed for January and February, shifting upward from the 30–40 ppb range to the 50 to 60 ppb range. This site had previously been identified as a location possibly influenced by Asian springtime long-range transport by Jaffe et al. (2003), Parrish et al. (2004), Jaffe and Ray (2007) and Oltmans et al. (2008), using various metrics (i.e., 8-h, daylight monthly averages, and changes in distribution patterns). For the period 1987–2006, Oltmans et al. (2008) report positive January, March, April, June, July, and

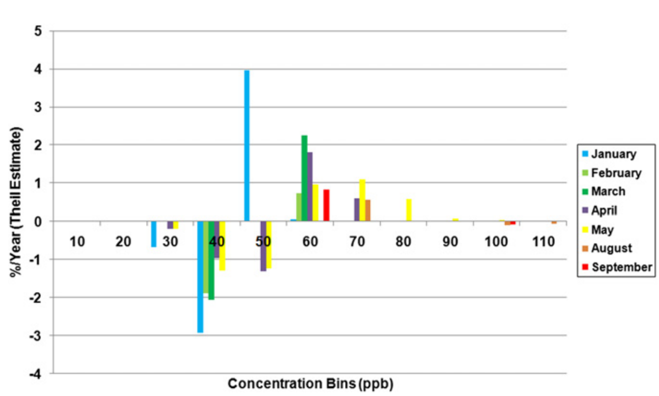

Fig. 10. Distribution of changes by month for a monitoring site located in Yosemite National Park (Turtleback Dome), California (AQS 060430003) for 1994–2008 for the months with significant changes.

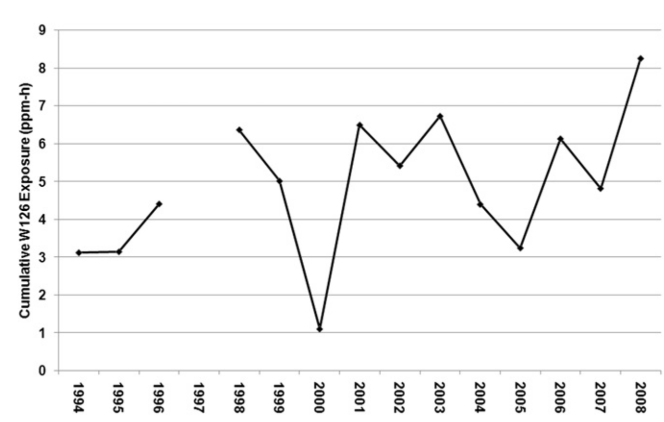

Fig. 12. Time series for the 24-h seasonal W126 exposure metric for Denali National Park (AK) (AQS 022900003) for 1994–2008. Incomplete data for 1997.

December trends. However, for 1994–2008 our current analysis indicates no statistically significant trends using the 3 exposure metrics and only wintertime trends in distributional shifts of the hourly values. The time period 1988–1993 exhibited low hourly O_3 with six of the lowest seven years over the entire 21-year record (1988–2008), while the year 1994 experienced twice the level of the average of the previous six years, contributing to the lack of trend in the most recent 15-year period. For 1994–2008 the data capture criterion was not met for calculating the 24-h W126 trend. However, by modifying the data capture criterion (i.e., by including years with two interpolations), we were able to investigate W126 metric trending patterns over a moving 15-year period. Although trending estimates (% per year) for 1988–2002, 1989–2003, 1990–2004, 1991–2005, 1992–2006, 1993–2007, and 1994–2008 decreased from the 1988–2002 period to the 1994–2008 period, none of the individual 15-year periods showed a statistically significant trend in the W126 metric because of the large variability that occurred over the period of record.

Although the year-to-year pattern for exposures was different at the Rocky Mountain NP (CO) site than at Lassen Volcanic NP, no trends were observed for either park for the metrics during 1994–2008. W126 trending patterns at Rocky Mountain NP over six moving 15-year periods (Fig. 13) yielded statistically significant positive trends during 1989–2003, 1990–2004, 1991–2005, 1992–2006, and 1993–2007. Because 2000, 2002, and 2003 experienced high 24-h W126 values in comparison to the other years, these years affected the positive trends for the earlier 15-year periods but not 1994–2008. Our results indicate that for Rocky Mountain NP it is important to investigate the change in the trending pattern with time (e.g., moving 15-year trending) in order to assess how year-to-year variability may influence the trend calculation. We noted that for Rocky Mountain NP, the W126 statistically significant positive trending patterns for five of the moving 15-year periods were affected by high exposure years in 2000, 2002, and 2003, while no trend was observed for the 15-year period 1994–2008.

Our results indicated that the quantification of the changes in the monthly hourly O_3 distributions provides supplemental information on trending in situations when the three exposure metrics used in our analysis did not provide statistically significant trends. Characterizing statistically significant monthly distributional changes in mid- and low-level hourly values is important for assessing physical processes associated with global climate change, long-range transport, and in modeling health risks that accumulate in concentration bins just above policy relevant background levels (US EPA, 2006). In addition, the quantitative assessment of the changes in the hourly distribution patterns as a function of emission reductions provides valuable information about the efficacy of the prospective control programs in reducing health and welfare risk (US EPA, 2006; Hazucha and Lefohn, 2007; Musselman et al., 2006; Heath et al., 2009). Careful selection of O_3 exposure metrics is necessary in these tasks because a statistically significant trend using one exposure index does not necessarily result in a similar trend using other metrics.

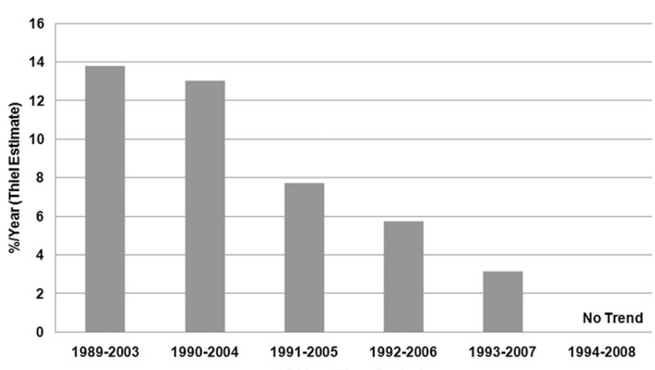

Fig. 13. Time Trends of 24-h W126 exposure metric over moving 15-year periods for Rocky Mountain National Park (AQS 080690007).

5. Conclusion

Using data for the periods 1980–2008 (29 years) and 1994–2008 (15 years), we explored whether historical trending patterns of surface O_3 at specific sites are continuing or changing. The majority of monitoring sites either (1) changed from negative (declining) to no trend, (2) remained negative, or (3) remained in the no trend status. Very few monitoring sites changed from either (1) no trend to negative or (2) no trend to positive (increasing) when compared over these periods. Approximately 45% of the monitoring sites shifted from negative to no trending status, indicating a leveling off for the 1980–2008 period. In characterizing the statistically significant changes in the distribution of hourly average O_3, we observed subtle statistically significant changes in the lower part of the distribution (i.e., below 50 ppb) that were not necessarily captured by the trending patterns associated with the three exposure metrics.

Using multisite data from 12 metropolitan cities, we found that as the frequency of higher hourly average concentrations was reduced, the lower hourly average concentrations also moved upward toward the mid-level values. The change in the number of the hourly average concentrations in the lower range is consistent with decreased NO scavenging. In assessing whether trending patterns are consistent across time periods, for Lassen Volcanic National Park (CA), trending estimates for 1988–2002, 1989–2003, 1990–2004, 1991–2005, 1992–2006, 1993–2007, and 1994–2008 were not significant. We attribute the non-significant trending to the large variability that occurred over the period of record. Using 6 moving 15-year time periods, we observed statistically significant positive trending patterns for 1989–2003, 1990–2004, 1991–2005, 1992–2006, and 1993–2007 at Rocky Mountain National Park (CO). No trends were observed for 1994–2008.

We recommend assessing possible subtle shifts in O_3 concentrations by characterizing changes in the distribution of hourly average concentrations by month. Identifying statistically significant monthly changes in the mid- and low-level hourly average concentrations may provide important information for assessing changes in physical processes associated with global climate change, long-range transport, and the efficacy of models used for emission and risk reductions. Careful selection of O_3 exposure metrics is necessary in these tasks because a statistically significant trend using one exposure index does not necessarily result in a similar trend using other metrics.

Appendix. Supplementary data

Supplementary data associated with this article can be found in the online version, at doi:10.1016/j.atmosenv.2010.08.049.

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