PERGAMON

Atmospheric Environment 35 (2001) 3217–3228

ATMOSPHERIC ENVIRONMENT

www.elsevier.com/locate/atmosenv

Trends in exceedances of the ozone air quality standard in the continental United States, 1980–1998

C.-Y. Cynthia Lin[1], Daniel J. Jacob*, Arlene M. Fiore

Division of Engineering and Applied Sciences and Department of Earth and Planetary Sciences, Harvard University, Pierce Hall, 29 Oxford St., Cambridge, MA 02138, USA

Received 24 September 2000; accepted 18 February 2001

Abstract

In 1997, the United States National Ambient Air Quality Standard (NAAQS) for ozone was revised from a 1-h average of 0.12 parts per million (ppm) to an 8-h average of 0.08 ppm. Analysis of ozone data for the ensemble of the contiguous United States and for the period 1980–1998 shows that the average number of summer days per year in exceedance of the new standard is in the range 8–24 in the Northeast and in Texas, and 12–73 in southern California. The probability of exceedance increases with temperature and exceeds 20% in the Northeast for daily maximum temperatures above 305 K. We present the results of several different approaches to analyzing the long-term trends in the old and new standards over the continental United States from 1980 to 1998. Daily temperature data are used to resolve meteorological variability and isolate the effects of changes in anthropogenic emissions. Significant negative trends are found in the Northeast urban corridor, in the Los Angeles Basin and on the western bank of Lake Michigan. Temperature segregation enhances the detection of negative trends. Positive trends occur at isolated sites, mostly in the Southeast; a strong positive trend is found in Nashville (Tennessee). There is some evidence that, except in the Southwest, air quality improvements from the 1980s to the 1990s have leveled off in the past decade.

Keywords: Air quality; Air quality standard; Ozone; Ozone trends; Ground-level ozone; Surface ozone

1. Introduction

High levels of surface ozone are responsible for most violations of the National Ambient Air Quality Standards (NAAQS) in the United States (US Environmental Protection Agency (EPA), 2000). Set in 1979, the original NAAQS for ozone was a 1-h average of 0.12 parts per million (ppm), not to be exceeded more than three times in 3 years. In July 1997, based on its review of scientific evidence linking ozone exposure to adverse health and welfare effects at levels allowed by the 1-h standard, the EPA revised the standard to an 8-h average of 0.08 ppm, with a form based on the 3-year average of the annual fourth-highest daily maximum 8-h average ozone concentrations measured at each monitor within an area (EPA, 1998). Because ozone concentrations are generally measured to the nearest part per billion (ppb), the smallest concentrations in exceedance of the old and new standards are 125 and 85 ppb, respectively. As of 1998, 51 million persons in the United States lived in areas violating the old standard, while 130 million persons lived in areas violating the new standard (EPA, 2000). In a study of rural sites in the eastern United States, Saylor et al. (1998) found that 2–12% of the sites violated the old standard and 30–50% of the sites violated the new standard for ozone during 1993–1995. A May 1999 federal appeals court ruling has

*Corresponding author. Tel.: +1-617-495-1794; fax: +1-617-495-4551.

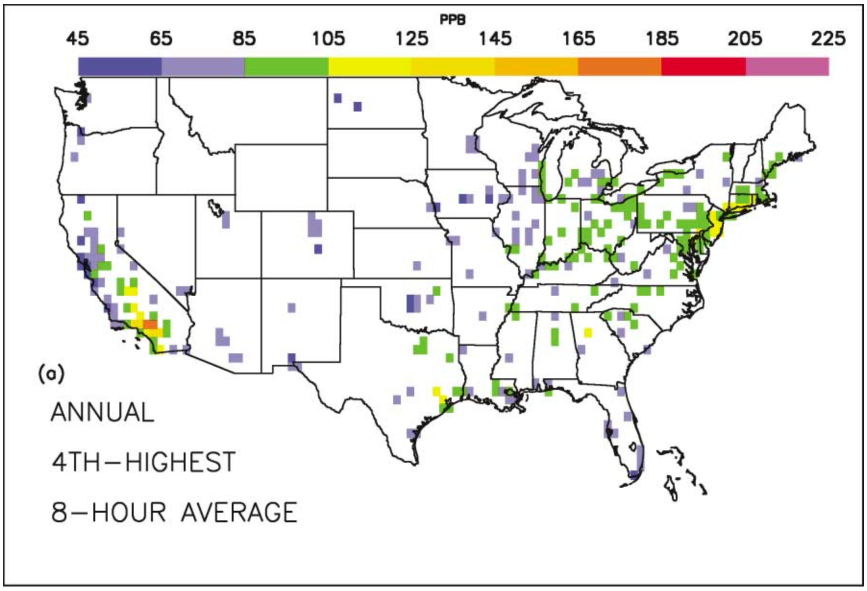
E-mail addresses: cclin@fas.harvard.edu (C.-Y.C. Lin), djj@io.harvard.edu (D.J. Jacob).

[1]Present address: Department of Economics, Harvard University, Cambridge, MA 02138, USA.

1352-2310/01/$ - see front matter
PII: S1352-2310(01)00152-2

questioned the constitutionality of the process by which the new standard was enacted; enforcement of this standard must wait until the issue is settled in the courts (American Trucking Associations vs. EPA, 1999).

We examine in this paper the long-term trends in exceedances of the new and old ozone standards in the United States over the past two decades. Considerable effort has been expended during this period to control anthropogenic emissions of ozone precursors, i.e., nitrogen oxides ($NO_x = NO + NO_2$) and hydrocarbons. These efforts have been hindered by population growth (31% from 1970 to 1997) and increased vehicle miles traveled (127% over the same period) (EPA, 1998). It is estimated that anthropogenic emissions of hydrocarbons in the United States decreased by 30% from 1980 to 1998, while NO_x emissions remained constant nationally to within a few percent (EPA, 1998, 2000). Hydrocarbon



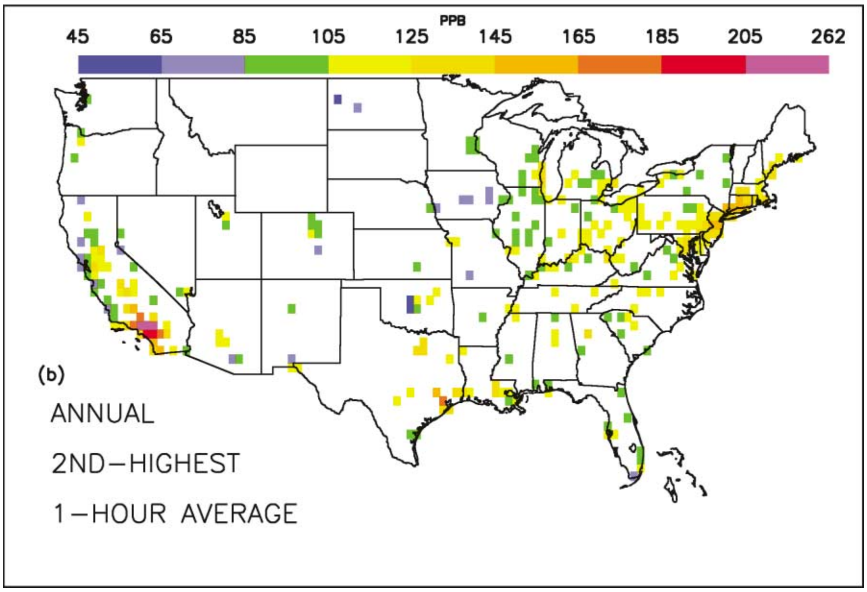


Fig. 1. Average values for 1980–1998 of (a) the annual fourth-highest daily maximum 8-h average ozone concentration (ppb) and (b) the annual second-highest daily maximum 1-h average ozone (ppb). The mean over all years is first calculated for each of the 522 individual sites meeting the data density requirement described in the text. The data are averaged for display over $0.5^\circ \times 0.5^\circ$ (roughly $50\,km \times 50\,km$) grid squares.

emission controls have been credited for abating the most extreme ozone pollution events, but a more general linkage between trends in precursor emissions and trends in ozone has yet to be demonstrated (National Research Council (NRC), 1991; Sillman, 1999; EPA, 2000).

Long-term trends or interannual variability of meteorological variables could contribute to or obfuscate ozone trends. Meteorological conditions influence ozone by affecting the biogenic emissions of hydrocarbons, the atmospheric circulation, and the photochemical environment (Rao et al., 2001). In particular, temperature is found to be strongly correlated with ozone concentrations on a day-to-day basis (Wolff and Lioy, 1978; Clark and Karl, 1982; NRC, 1991). Higher temperatures enhance biogenic hydrocarbon emissions and increase the abundance of NO_x by thermal decomposition of the peroxyacetyl nitrate (PAN) reservoir (Olszyna et al., 1997). Temperature is also a surrogate for clear skies and stagnation (Jacob et al., 1993; Olszyna et al., 1997; Smith and Adamski, 1998). As a result, it is found that the correlation with temperature can account for most of the influence of meteorological variables on ozone (Jacob et al., 1993).

Many previous studies have examined long-term trends of ozone concentrations in selected regions of the United States (Rao et al., 2001, and references therein). In an examination of nationwide trends in medians and 90th percentiles of summer afternoon ozone for 1980–1995, Fiore et al. (1998) found that trends were insignificant over most of the continental United States; decreasing trends were clustered in the New York City, Los Angeles and Chicago metropolitan areas. We extend here on the above studies by presenting a trend analysis targeted at exceedances of both the 1- and 8-h ozone standards for the entire contiguous United States and for nearly two decades (1980–1998). We also attempt to isolate the contributions of anthropogenic emissions to the trends by using temperature as a proxy for meteorological influences.

2. Data and methods

Hourly ozone concentration data from 1980 to 1998 were extracted from the EPA's Aerometric Information Retrieval System (AIRS). Ozone measurements at the AIRS sites are subject to frequent calibration and consistency checks. We chose to begin the analysis in 1980, a year after the EPA used an ultraviolet photometric method to uniformly calibrate all ozone measurements (Chock, 1989). The AIRS database includes over 2000 sites across the United States that have monitored ozone at one time or another over the 19 years of our data set, but many of these sites have sparse records. For cases in which data for sequential years were available from sites a few kilometers apart, usually because the monitoring station had moved, the ozone data were merged.

In accordance with the protocol for the new standard (EPA, 1997), running 8-h averages, indexed by the first hour, were calculated for each 8-h interval with at least 6 h of data, and the daily maximum 8-h average was stored for each day for each site. Similarly, daily maximum 1-h average ozone concentrations were also stored for each day for each site.

Fig. 1 shows the average values over the 19 years of the data set of the annual fourth-highest daily maximum 8-h average ozone and the annual second-highest daily maximum 1-h average ozone for sites with at least 15 years fulfilling the minimum data density requirement of 4 and 2 days per year, respectively. For both the 8-h and 1-h analyses, there were 522 sites that fulfilled the data density requirement. The 19-year mean was calculated for each individual site and then averaged for display over 0.5° × 0.5° (roughly 50 km × 50 km) grid squares. For both statistics, the highest values are in southern California, along the Northeast coast, in Atlanta, and in Houston. More sites are in violation of the new 8-h standard, as denoted by the green, yellow,

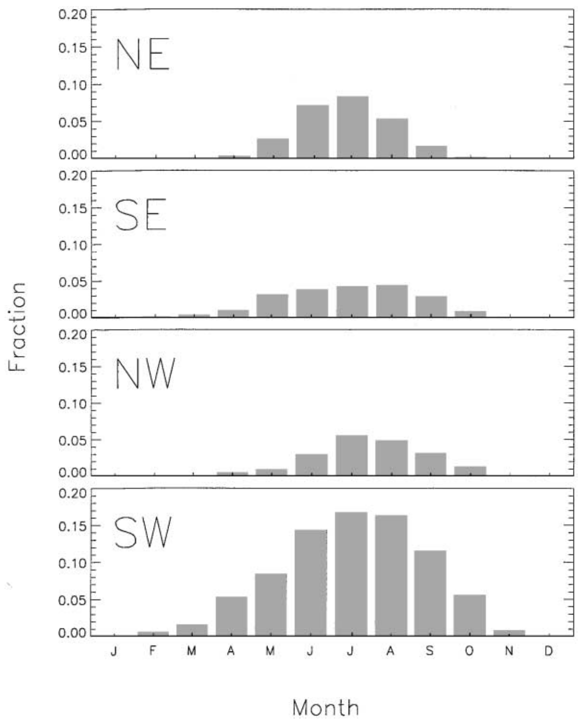


Fig. 2. Average fraction of days per month whose daily maximum 8-h average exceeds 85 ppb, out of all days per month with ozone data, for the analysis period 1980–1998 and for four quadrants of the United States partitioned at 36°N, 97.5°W (northern Oklahoma).

orange, and red squares, than are in violation of the old 1-h standard, as denoted by the dark yellow, orange and red squares.

In addition to rank order, an alternate type of statistic that can be used to analyze exceedances of the ozone air quality standard is the frequency of occurrence of daily maximum 1- or 8-h average concentrations in excess of the standards. Seasonal selection of the data is appropriate here to avoid any bias caused by site-to-site differences in the yearly extent of data coverage; most sites do not operate year-round. Fig. 2 shows the average fraction of days in exceedance of the 8-h standard by month for the four quadrants of the United States, as partitioned at 36°N, 97.5°W (in northern Oklahoma). For each month and for each year, the fraction of exceedance days out of all

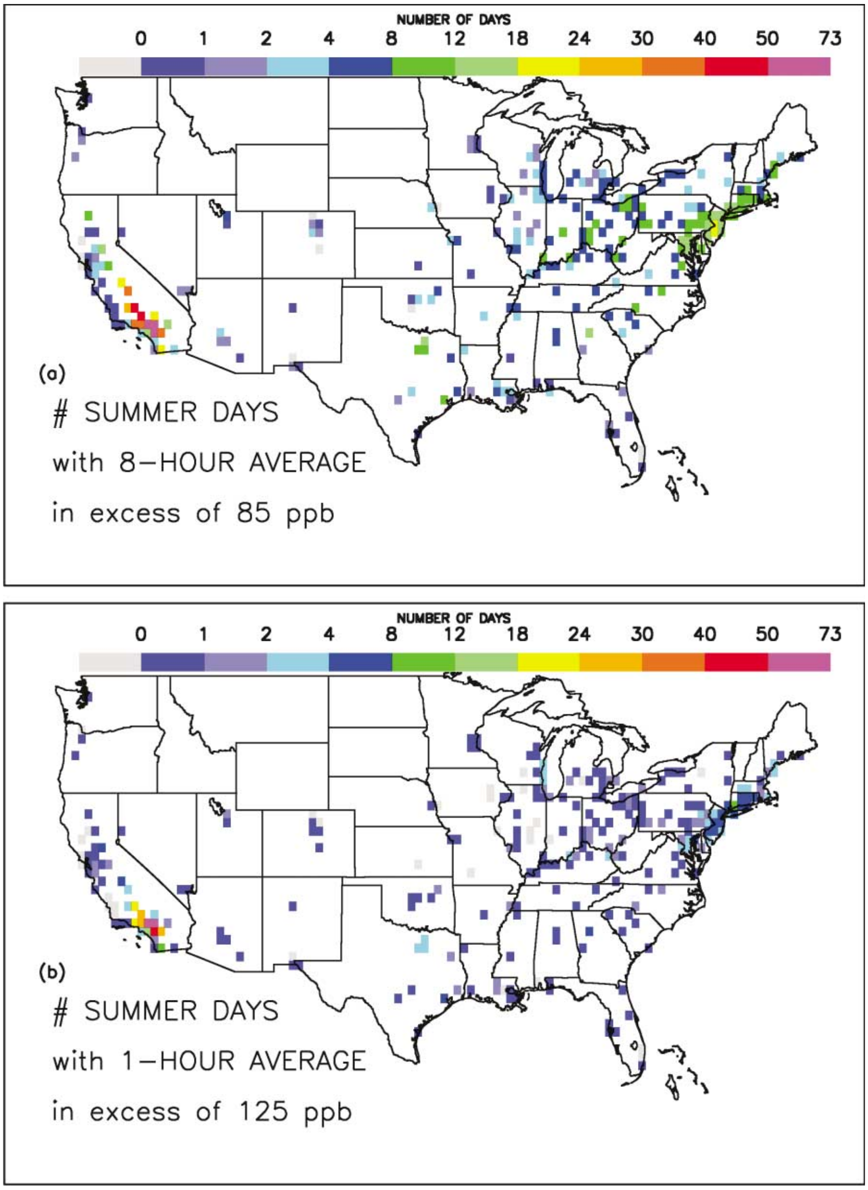


Fig. 3. Average number of summer days per year (a) whose daily maximum 8-h average exceeds 85 ppb and (b) whose daily maximum 1-h average exceeds 125 ppb. The analysis period is 1980–1998. The mean over all years is first calculated for each individual site meeting the data density requirement described in the text. The data are averaged for display over 0.5° × 0.5° grid squares. A total of 462 sites were used for (a); 463 sites were used for (b).

days with data was calculated for each site; the resulting values for each month were then averaged across all years and for all sites in each quadrant. We find that the summer months (June–August) account for 81% of all the exceedances in the Northeast, 60% in the Southeast, 70% in the Northwest, and 58% in the Southwest.

We therefore focus our trend analysis of the fraction of summer days in exceedance of the new and old standards, and require that sites have at least 16 days of data per month for all three summer months and for at least 15 of the 19 summers. For ease of comprehension, the fractions are expressed as the number of summer days in exceedance per year via multiplication by 92. Figs. 3a and b show the average values of the number of summer days in exceedance of the 8- and 1-h standards over 1980–1998, respectively. The greatest frequency of exceedances is in

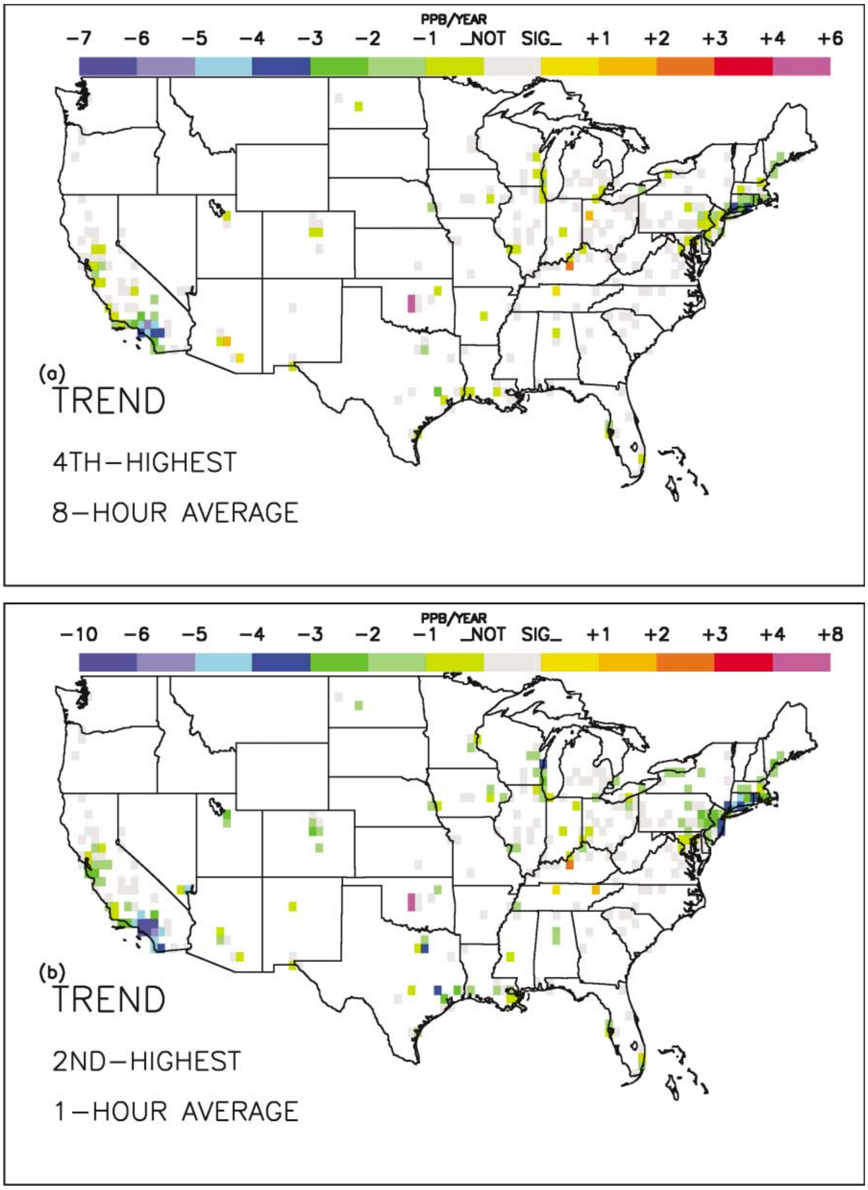


Fig. 4. Linear trend in ppb/year over 1980–1998 of (a) the annual fourth-highest daily maximum 8-h average ozone, and (b) the annual second-highest daily maximum 1-h average ozone. Trends are calculated for individual sites and then averaged for display over $0.5^\circ \times 0.5^\circ$ grid squares. Sites with statistically insignificant trends enter this averaging as a trend of zero. Grid squares whose average trend is zero and therefore insignificant are shown in gray.

southern California, where exceedances occur 12–73 times per summer for both standards. Many exceedances also occur in the Northeast and eastern Texas (8–24 and 2–8 occurrences per summer for the 8- and 1-h standards, respectively). In general, sites experience more exceedances of the 8-hour standard than of the 1-h standard.

3. Results

3.1. Trends in exceedances of the ozone standard

Fig. 4 shows the 1980–1998 trends in the annual fourth-highest daily maximum 8-h average ozone concentration and the annual second-highest daily

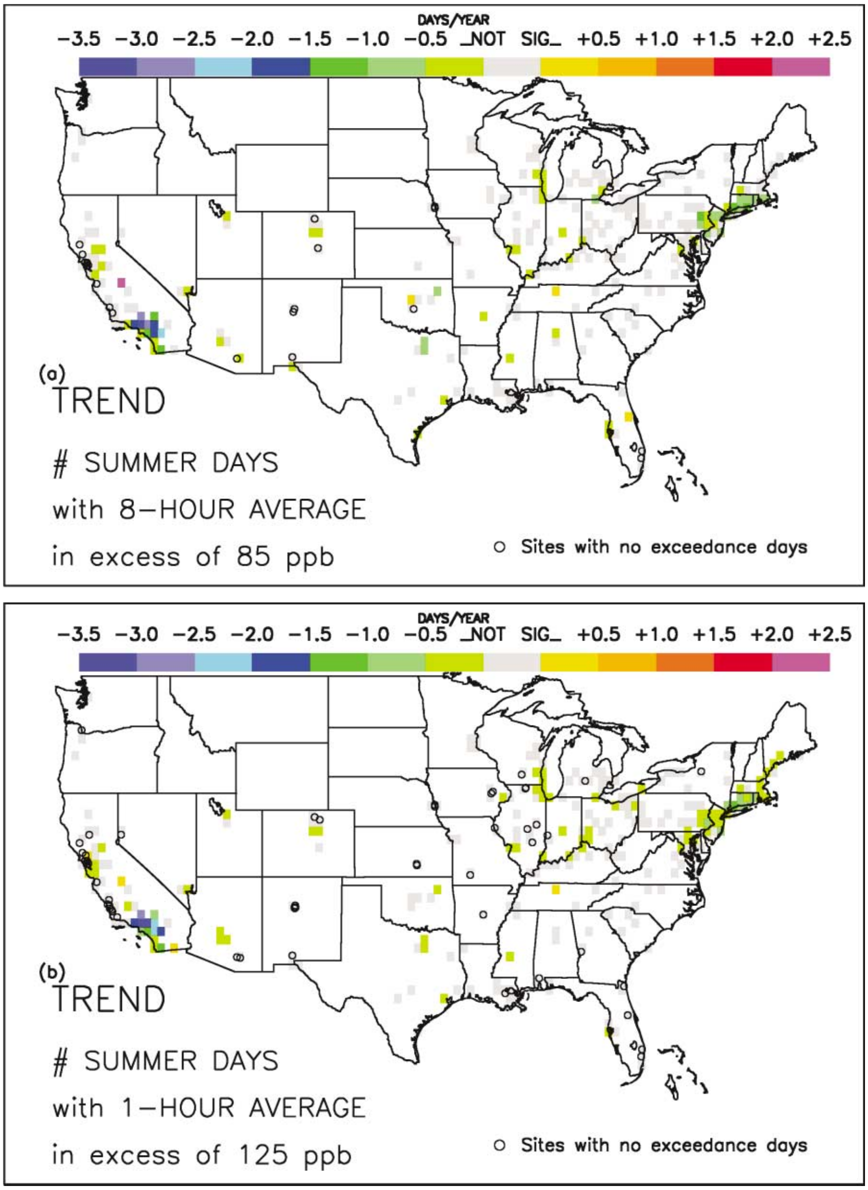


Fig. 5. Linear trend in days/year over 1980–1998 in the number of days per summer (a) whose daily maximum 8-h average exceeds 85 ppb and (b) whose daily maximum 1-h average exceeds 125 ppb. Trends are calculated for individual sites and then averaged for display over $0.5^\circ \times 0.5^\circ$ grid squares. Sites with statistically insignificant trends enter the calculation as a trend of zero. Grid squares whose average trend is zero and therefore insignificant are shown in gray. Circles indicate sites with no exceedance days for any of the summers over the 1980–1998 period.

maximum 1-h average ozone concentration, for the ensemble of sites shown in Fig. 1. A linear regression is conducted to detect trends that are statistically significant at a 5% level. Trends are calculated for individual sites and then averaged for display over 0.5° × 0.5° grid squares. Sites with statistically insignificant trends enter the averaging as a trend of zero. Grid squares whose average trend is zero and therefore insignificant are

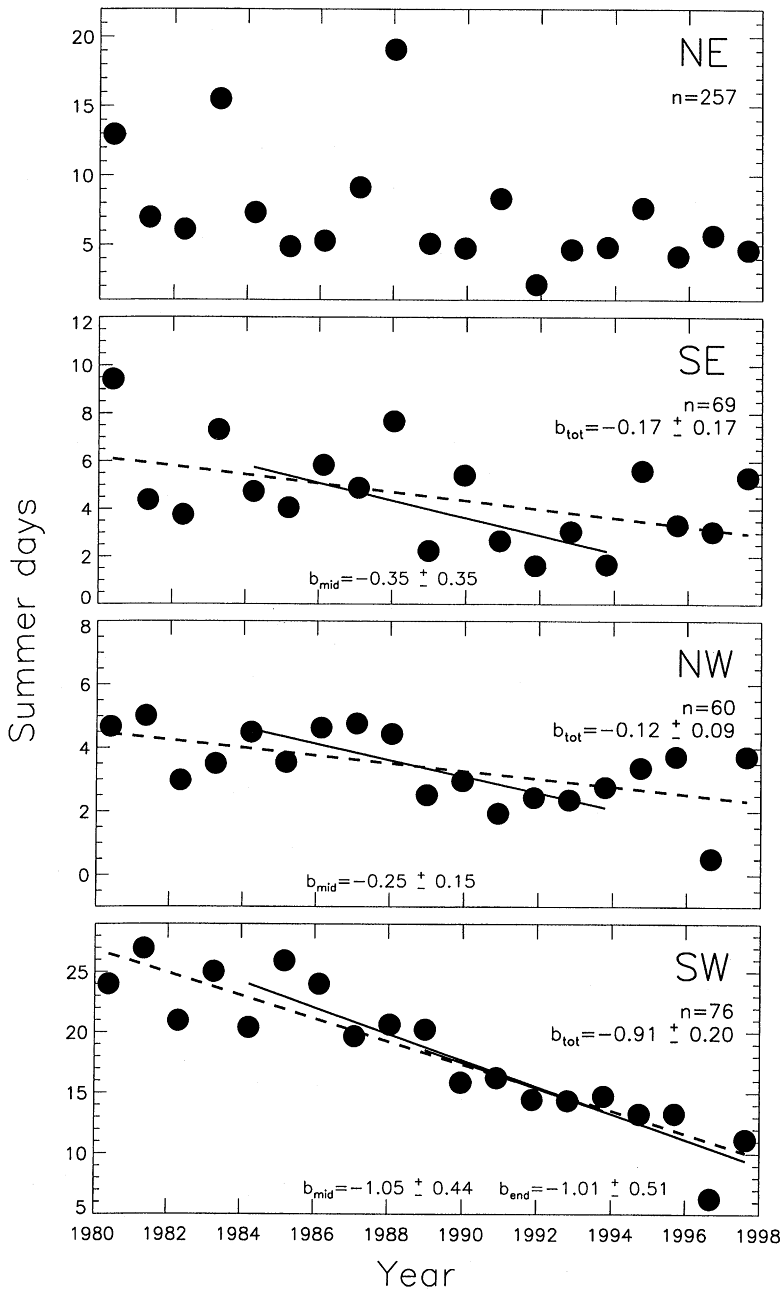


Fig. 6. Time series of the number of summer days per year whose daily maximum 8-h average exceeds 85 ppb, averaged over all sites meeting the data density requirement for each quadrant of the United States over 1980–1998. The quadrants are partitioned at 36°N, 97.5°W. Significant trends for the 19-year period are denoted by a dashed line; slopes b_{tot} and confidence intervals are given in units of days per year. Similarly, significant trends for 1980–1989, 1984–1994, and 1989–1998 are denoted by solid lines; their respective slopes b_{beg}, b_{mid} and b_{end} and confidence intervals are given in units of days per year.

shown in gray. For the 8-h statistic, the strongest downwards trends occur along the Northeast and Southwest coasts, especially in southern Connecticut and southern California; localized increasing trends occur in Ohio, Kentucky, Tennessee, Oklahoma, and Arizona. More sites have significant negative trends in the 1-h statistic than in the 8-h statistic and the trends are larger. Only two sites in Tennessee, one site in Kentucky, and a few sites in Oklahoma have significant positive trends in the 1-h statistic.

Fig. 5 shows the trends for 1980–1998 in the number of summer days per year in exceedance of the 8-h standard (85 ppb) and the 1-h standard (125 ppb). The trends were calculated using linear regression as discussed above. Sites at which there have been no exceedance days in any of the summers are indicated

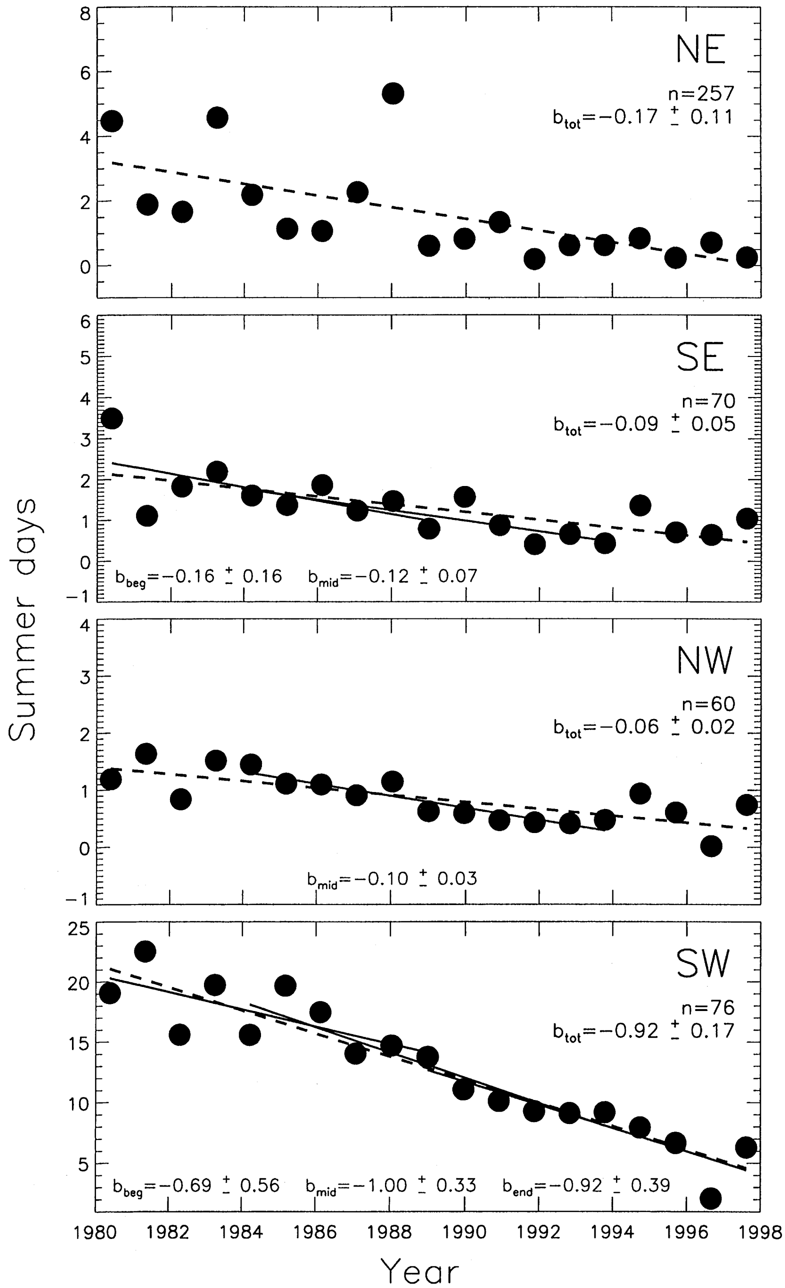


Fig. 7. Same as Fig. 6 but for the number of summer days per year whose daily maximum 1-h average exceeds 125 ppb.

with an open circle. The strongest downward trends are in the Los Angeles Basin; strong downward trends also occur in the Northeast urban corridor. Significant positive trends in the 8-h standard occur in Tennessee, Oklahoma, Florida, and central California, but are isolated. The only significant positive trends in the 1-h standard are in Tennessee and at a few sites in California. In Fresno (California), there is a large significant positive trend in the number of summer days in exceedance of the new standard but no significant trend in the fourth-highest concentration.

Strongly nonlinear ozone trends might not be detected by a linear regression. As an alternate approach to diagnosing the trends, we compared ozone statistics for the first 5 years (1980–1984) and the last 5 years (1994–1998) of the record. Details of this analysis are reported by Lin (2000) and a summary of results for the ensemble of the United States is presented by Lin et al. (2000). The results of the comparison between the first and last 5 years for the 8-h standard are similar to those of our linear trend analysis (Fig. 5a). For the 1-h standard, many of the sites which have significant negative linear trends (Fig. 5b) do not have significant changes from the first to the last 5 years.

A perspective on the linearity of the trends over the 1980–1998 period is presented in Figs. 6 and 7, which show the time series of the average number of days in exceedance of the 8-h standard and 1-h standard, respectively, for each quadrant of the United States partitioned at 36°N, 97.5°W. For each year, the fraction of exceedance days for each site meeting the data density requirement is averaged over all sites in each quadrant. Significant negative linear trends are found for all quadrants for both standards, except for exceedances of the 8-h standard in the Northeast. The trend for the 8-h standard in the Northeast becomes significant if 1988 is removed from the regression ($b = -0.30 \pm 0.25$ days/year). To investigate possible variations in trends, we conducted linear trend analysis on the first 10 years (1980–1989), middle 11 years (1984–1994), and last 10 years (1989–1998) of the time series. For the 8-h standard (Fig. 6), trends for the middle 11 years are significant for all quadrants except the Northeast, while trends for the first 10 years and last 10 years are insignificant for all quadrants except the Southwest, which has a significant negative trend for the last 10 years. For the 1-h standard (Fig. 7), the Southeast has significant negative trends for both the first 10 years and the middle 11 years, the Northwest has significant negative trends for the middle 11 years, and the Southwest has significant negative trends for all three intervals. Except in the Southwest, the number of summer days per year in exceedance of either standard has not changed significantly over the last 10 years of the data set for any quadrant. This would suggest that air quality gains from the 1980s to the 1990s have not continued in the past decade.

3.2. Segregation by temperature

Segregation of the ozone data by temperature can be used to filter the influence of meteorological variables on the trend and isolate the effect of changes in ozone concentrations due to changes in anthropogenic emissions (Fiore et al., 1998; Rao et al., 2001). For this purpose we use time series of daily maximum temperature data from National Climatic Data Center (NCDC) sites. Following Fiore et al. (1998), one representative NCDC site was selected for each 4° latitude by 5° longitude grid square. A finer resolution would be inappropriate because of the long lifetime of ozone.

We find no significant long-term trends in temperature over the 1980–1998 period for any of our NCDC sites, but there is interannual variability (Lin, 2000). For example, the summer of 1988 was unusually hot

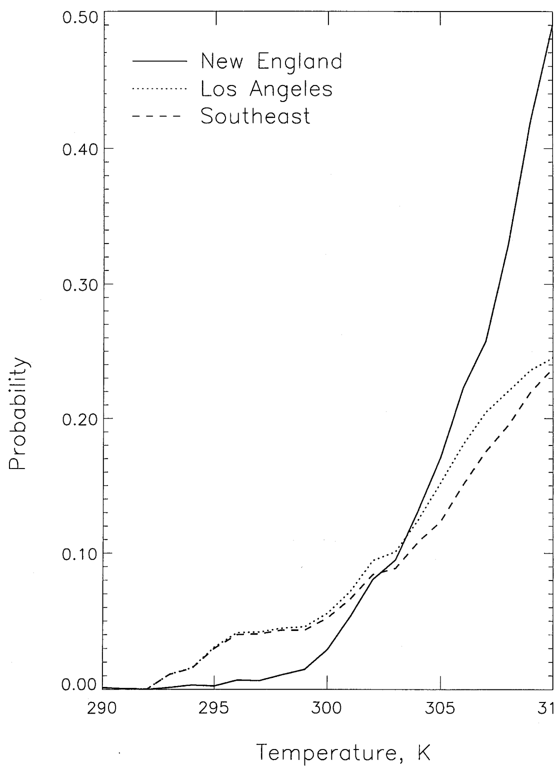


Fig. 8. Probability that the daily maximum 8-h average ozone will exceed 85 ppb for a given daily maximum temperature, based on 1980–1998 data. Values are shown for New England (bounded by 36°N, 44°N, 67.5°W, and 87.5°W), the Los Angeles basin (bounded by 32°N, 40°N, 112.5°W, and 122.5°W) and part of the Southeast (bounded by 32°N, 36°N, 72.5°W, and 92.5°W).

in most of the eastern United States, and ozone concentrations were especially high that year (Figs. 6 and 7; also see Baumgardner and Edgerton, 1998). The year of 1995 has also been cited as a warm summer (Chock et al., 1997).

To investigate the relationship between temperature and ozone exceedances, we calculate the probability that an ozone exceedance will occur when the daily maximum temperature is in a given range. For each temperature bin of width 1 K, we determine the fraction of site-days whose daily maximum temperature fell into that bin and the fraction of those days whose daily maximum 8-h ozone exceeded the standard. We define one site-day to be 1 day in summer for which ozone data is available for one site; two site-days might therefore consist of two different days for the same site, the same day for two different sites, or two different days for two different sites. Fig. 8 displays the result of this analysis for New England, the Los Angeles basin and part of the Southeast. As expected, the probability of exceedance increases with temperature. Exceedance probabilities for a temperatures above 303 K tend to be higher in the Northeast than in other parts of the United States, reaching 49% at 310 K in New England. However, such high temperatures are rare in that region.

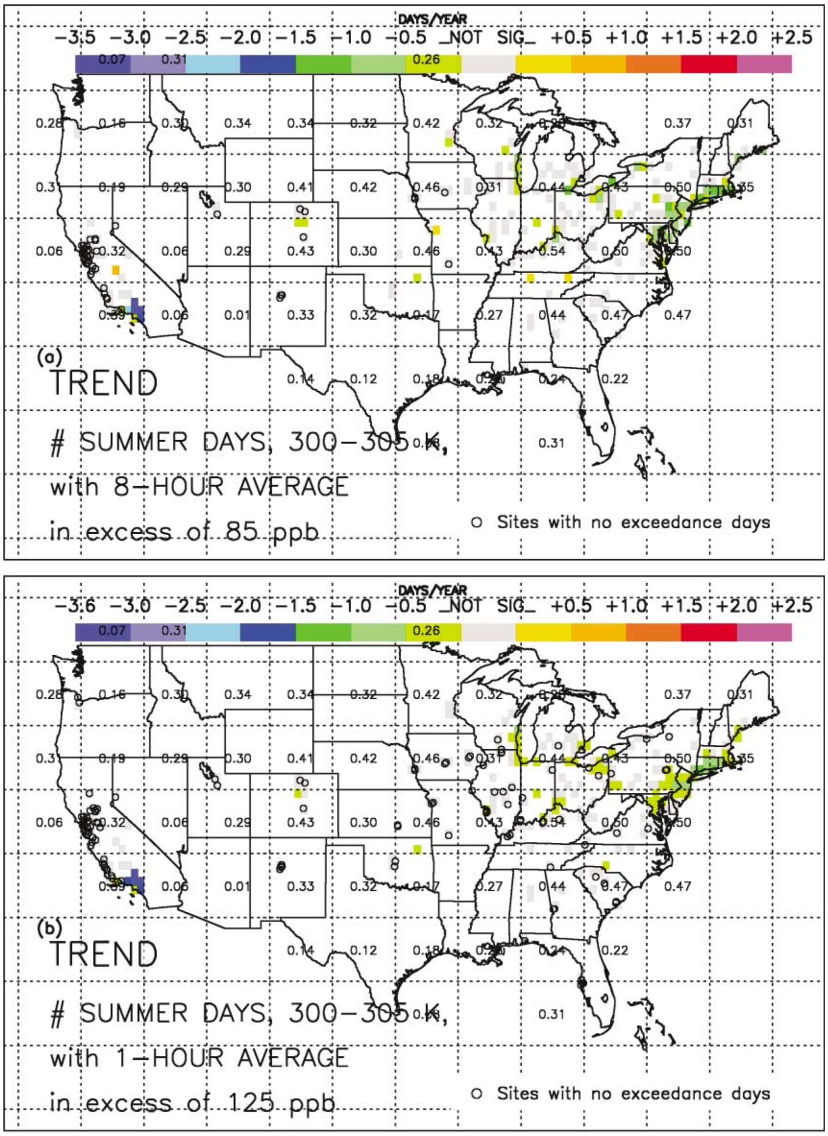


Fig. 9. Same as Fig. 5 but for days when the maximum temperature fell in the range 300–305 K. The average fraction of summer days falling in this temperature bin is shown for each 4° latitude by 5° longitude grid square.

We repeat the trend analyses described in Section 3.1 after restricting the data to only those days whose daily maximum temperature fell between 300 and 305 K, inclusive. A 300–305 K temperature bin has previously been used in trend analyses conducted by Fiore et al. (1998). Fig. 9 presents results from the same trend analysis used for Fig. 5, but with segregation by temperature. The data density requirement was relaxed to a minimum of 16 days per summer rather than 16 days per month. Compared to Fig. 5, temperature binning allows detection of significant negative trends in the 8-h average statistic at many more sites along the East Coast and along the Great Lakes. Other features include a stronger negative trend in Connecticut, at Los Angeles sites and along the Great Lakes; significant positive trends in Tennessee and central California as well as in Missouri; and removal of the significant positive trends in Oklahoma and in Florida. For the 1-h average statistic, temperature binning reveals more significant negative trends in Pennsylvania, eliminates the significant positive trend in Tennessee and removes some of the positive and negative trends in California.

4. Conclusions

This paper presented the results of several different approaches to analyzing trends in exceedances of ozone air quality standards over the continental United States for the 1980–1998 period, for both the old standard (125 ppb, 1-h average) and the new standard (85 ppb, 8-h average). Exceedances are considerably more widespread for the new standard than for the old standard. Although significances and magnitudes vary, downward trends in southern New England, along the western bank of Lake Michigan, and in the Los Angeles Basin are detected by all trend analyses. Upward trends are seen only at isolated sites which vary depending on the analysis, except for Nashville (Tennessee), where a significant upward trend appears in all analyses except that of the 1-h standard with temperature binning (Fig. 9b). In general, binning the data by temperature to remove the effect of interannual variability in weather reveals stronger and more significant downward trends.

In general, downward trends are greater and more significant for the old standard than for the new standard, but there is a close correspondence between both standards in the regions where significant downward trends have been achieved. It therefore appears that emission control policies enacted to bring areas into compliance with the old standard have also been effective in lowering exceedances of the new standard.

Except for the Southwest, quadrants of the United States that experienced significant downward trends in ozone exceedances over the 19-year period did not experience significant downward trends over the last decade (1989–1998), indicating that improvements in air quality from the 1980s to the 1990s may have leveled over the course of the 1990s.

Acknowledgements

This work was supported by an undergraduate research fellowship from the Harvard College Research Program to C.-Y.C. Lin, and by the National Science Foundation.

References

American Trucking Associations, Inc., et al., 1999. United States Environmental Protection Agency (No. 97-1440) (Online: web). URL: http://www.epa.gov/ttn/oarpg/gen/97-1440a.txt.

Baumgardner, R.E., Edgerton, E.S., 1998. Rural ozone across the eastern United States: analysis of CASTNet data, 1988–1995. Journal of Air and Waste Management Association 48, 674–688.

Chock, D.P., 1989. The need for a more robust ozone air quality standard. Journal of Air Pollution Control Association 39, 1063–1072.

Chock, D.P., Winkler, S.L., Pezda, S., 1997. Attainment flip-flops and the achievability of the ozone air quality standard. Journal of Air Waste Management Association 47, 620–622.

Clark, T.L., Karl, T.R., 1982. Application of prognostic meteorological variables to forecasts of daily maximum one hour ozone concentrations in the northeastern United States. Journal of Applied Meteorology 21, 1662–1671.

Environmental Protection Agency (EPA), 1997. National ambient air quality standards for ozone; final rule (40 CFR Part 50). Federal Register 62 (138).

Environmental Protection Agency (EPA), 1998. National air quality and emission trends report, 1997 (EPA Report No. EPA 454/R-98-016). Research Triangle Park, NC.

Environmental Protection Agency (EPA), 2000. National air quality and emission trends report, 1998 (EPA Report No. EPA 454/R-00-003). Research Triangle Park, NC.

Fiore, A.M, Jacob, D.J., Logan, J.A., Yin, J.H., 1998. Long-term trends in ground level ozone over the contiguous United States, 1980–1995. Journal of Geophysical Reserach 103 (D1), 1471–1480.

Jacob, D.J., Logan, J.A., Gardner, G.M., Yevich, R.M., Spivakovsky, C.M., Wofsy, S.C., Sillman, S., Prather, M.J., 1993. Factors regulating ozone over the United States and its export to the global atmosphere. Journal of Geophysical Research 98 (D8), 14817–14826.

Lin, C.-Y.C., 2000. Trends in ozone smog. Senior Thesis, Harvard University.

Lin, C.-Y.C., Jacob, D.J., Munger, J.W., Fiore, A.M., 2000. Increasing background ozone in surface air over the United States. Geophysical Research Letters 27 (21), 3465–3468.

National Research Council (NRC), 1991. Rethinking the Ozone Problem in Urban and Regional Air Pollution. National Academy Press, Washington, DC.

Olszyna, K.J., Luria, M., Meagher, J.F., 1997. The correlation of temperature and rural ozone levels in southeastern U.S.A. Atmospheric Environment 31 (18), 3011–3022.

Rao, S.T., Porter, P.S., Zurbenko, I.G., Dunker, A.M., & Wolff, G.T., 2001. Ozone air quality over North America: a critical review of trend detection techniques and assessments. Journal of Air and Waste Management Association (in press).

Saylor, R.D., Chameides, W.L., Cowling, E.B., 1998. Implications of the new National Ambient Air Quality Standards for compliance in rural areas. Journal of Geophysical Research 103 (D23), 31137–31141.

Sillman, S., 1999. The relation between ozone, NO_x and hydrocarbons in urban and polluted rural environments. Atmospheric Environment 33, 1821–1845.

Smith, B.E., Adamski, W.J., 1998. Eight-hour ozone trends at sites in Lake Michigan ozone nonattainment areas. Journal of Air Waste Management Association 48, 1204–1206.

Wolff, G.T., Lioy, P.J., 1978. An empirical model for forecasting maximum daily ozone levels in the northeastern U.S. Journal of Air Pollution Control Association 28, 1034–1038.