JOURNAL OF GEOPHYSICAL RESEARCH, VOL. 103, NO. D1, PAGES 1471-1480, JANUARY 20, 1998

Long-term trends in ground level ozone over the contiguous United States, 1980–1995

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Abstract. Long-term trends of median and 90th percentile summer afternoon O_3 concentrations were examined at 549 sites across the United States for the 1980-1995 period. Daily temperature data were used to account for the variability in O_3 concentrations associated with temperature. Both before and after segregating the O_3 data by temperature, trends were insignificant over most of the continental United States. No region of the United States experienced a significant increase in O_3 concentrations during the 1980-1995 period. Decreasing trends were predominantly clustered in the three largest metropolitan areas: New York City, Los Angeles, and Chicago. In these areas, additional sites with trends were identified in the temperature-segregated analysis. Correlation of trends with local anthropogenic emissions of nitrogen oxides ($NO_x = NO + NO_2$) and volatile organic compounds (VOC) indicates a greater frequency of decreasing trends for urban sites with high emission. National emission inventories for the United States indicate that anthropogenic VOC emissions decreased by 12% over the 1980-1995 period while NO_x emissions remained constant. The observed O_3 trends are consistent with the view that summertime O_3 production over the United States is NO_x-limited except in the largest metropolitan areas where it is partly VOC-limited.

1. Introduction

Ground level O_3 "smog" is produced from complex photochemical reactions involving nitrogen oxides (NO_x) and volatile organic compounds (VOCs). Ozone in surface air is hazardous to human lungs and has been blamed for billions of dollars in agricultural damage in the United States [*Prinz*, 1988; *Lippmann*, 1991]. As of 1995, approximately 71 million Americans lived in counties unable to meet the National Ambient Air Quality Standard (NAAQS) for O_3 set by the United States Environmental Protection Agency (EPA) at a 1-hour average of 120 parts per billion by volume (ppb) not to be exceeded more than 3 times in three years [*EPA*, 1996a]. The NAAQS standard was revised in July, 1997 to an 8-hour average of 80 ppb, which is expected to increase the extent of noncompliance [*Chameides et al.*, 1997].

Over the past 25 years, substantial effort has been invested in controlling precursor emissions with the aim of reducing O_3 levels. The primary focus has been on controlling VOCs [*Calvert et al.*, 1993; *Tietenberg*, 1996]. Between 1980 and 1995, anthropogenic VOC emissions in the United States decreased by 12%, while NO_x emissions remained constant, even though the annual number of vehicle kilometers driven rose by 60% [*EPA*, 1996b]. Recent modeling studies have indicated that over most of the United States, O_3 production is limited by the supply of NO_x rather than VOCs [*Trainer et al.*, 1987; *Sillman et al.*, 1990a; *McKeen et al.*, 1991a,b; *National Research Council (NRC)*, 1991; *Chameides et al.*, 1992; *Jacob et al.*, 1993]. The 1990 Clean Air Act Amendments imposed stricter NO_x controls on new cars and power plants, but these changes will impact national NO_x emissions only gradually.

A number of studies have examined long-term trends of O_3 concentrations in selected regions of the United States (Table 1). Most of the focus has been on the northeastern United States and southern California, which frequently exceed the NAAQS [*EPA*, 1996a]. Methods have ranged from ordinary least squares regression analysis [e.g., *Walker*, 1985] to complex regression models that account for the effects of multiple meteorological variables on O_3 [e.g., *Flaum et al.*, 1996]. Ozone concentrations are strongly correlated with temperature on a day-to-day basis [*Wolff and Lioy*, 1978; *Clark and Karl*, 1982; *NRC*, 1991], and this correlation is generally accounted for in O_3 trend analyses (Table 1). Some of the studies in Table 1 based their diagnostics of trends on the 1-hour daily maximum O_3 concentration, while others focused on extreme yearly statistics to reflect the formulation of the NAAQS. Most recently, the EPA reported a 6% decrease from 1986 to 1995 in the national average of daily maximum 1-hour O_3 concentrations, and a 53% decrease in the number of exceedances of the NAAQS [*EPA*, 1996a].

Our work expands upon the previous studies in Table 1 by examining the spatial distribution of trends on a national scale, using 1980-1995 data from all O_3 monitoring sites included in the EPA Aeronometric Information Retrieval Service (AIRS). The analysis begins in 1980 to avoid difficulties with instrument calibrations; beginning in 1979, the AIRS sites have used the ultraviolet photometric method to uniformly calibrate all O_3 measurements, facilitating comparisons among sites [*Chock*, 1989]. We focus on the medians and 90th percentiles of summer afternoon O_3 concentrations, as these statistics are more robust diagnostics of O_3 than extrema. Daily temperature data are used to account for the correla-

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Paper number 97JD03036.
0148-0227/98/97JD-03036$09.00

Table 1. Studies of Summertime Ozone Trends in the United States

Reference	Time period Analyzed	Statistics Used	Region/Sites	Method of Analysis	Meteorological Variables Factored Into Analysis[a]
Walker [1985]	1973-1982 (1975-1982 for Texas)	annual mean, annual hours in exceedance of NAAQS, mean daily max	California: 50 sites; Texas: 15 sites	linear regression	none
Kuntasal and Chang [1987]	1968-1985	multistation mean of daily 1-hour max	SCAB[b]: nine sites	linear regression	T
Zeldin et al. [1990]	1981-1989	daily 1-hour max, days exceeding 200 ppb	SCAB: eight station pairs	linear regression	24 standardized variables
Cassmassi and Bassett [1991]	1976-1990	hours > 120 ppb and hours > 200 ppb	SCAB: mean of all sites	linear regression	T, precipitation, WS, sky cover
Korsog and Wolff [1991]	1973-1983	75th percentile of daily 1-hour max	northeastern United States: eight sites	linear regression[c]	T, RH, WS
Lefohn and Shadwick [1991]	1979-1988	hourly averages weighted by exposure indices[d]	77 rural sites nationwide	linear regression[e]	None
Rao et al. [1992]	1980-1989	median of daily 1-hour max	New Jersey, New York, and Connecticut	various statistical methods[f]	None
Cox and Chu [1993]	1981-1991	daily 1-hour max	43 urban areas exceeding NAAQS	probability model: Weibull distribution	T, WS, RH, opaque cloud cover, mixing height
Rao et al. [1995]	1983-1992	daily 1-hour max	eastern United States	K-Z filter[g]	T
Zurbenko et al. [1995]	1980-1992	daily 1-hour max	eastern United States	K-Z filter	T
Flaum et al. [1996]	1983-1992	daily 1-hour max	eight eastern United States sites	K-Z filter	T, RH
EPA [1996a, b]	1986-1995	second daily 1-hour max	183 U.S. MSAs[h]	linear regression	None
This study	1980-1995	summer afternoon median and 90th percentile	549 sites nationwide	linear regression	T

[a]T, temperature; RH, relative humidity; WS, wind speed.

[b]SCAB, South Coast Air Basin of California.

[c]Used fixed-value and fixed-range criteria.

[d]Exposure indices weigh the concentrations according to their adverse affects on vegetation.

[e]Determined significance of slope by the Mann-Kendall test for trend.

[f]The bootstrap method, extreme value statistics, and the Mann-Kendall test for trend.

[g]The Kolmogorov-Zurbenko filter is first applied by *Rao et al.* [1994]. It consists of repeated iterations of a simple moving average. It separates the trend component from the seasonal and short-term variation components of a time series consisting of the logarithm of daily maximum O_3 concentrations.

[h]MSAs, Metropolitan Statistical Areas.

tion of O_3 with temperature. Relationships of trends with local anthropogenic emissions of NO_x and VOC are also examined.

2. Data and Methods

Records of hourly O_3 concentrations for 1980-1995 were obtained from EPA for all AIRS stations in the continental United States (approximately 900 sites). Many of the AIRS sites are clustered around major cities; rural regions have few monitoring stations. The density of sites is greater in the east than in the west (with the exception of California). Our analysis focuses on summer afternoons (June through August), when O_3 is at its seasonal and diurnal maximum [*Logan*, 1989]. For each summer day in the 1980-1995 record, hourly concentrations from 13 to 16 local time (LT) were extracted. Medians and 90th percentiles of these concentrations are displayed in Plate 1. Much of the eastern and southwestern United States have median concentrations greater than 50 ppb. Concentrations are generally lower along the west coast than along the east coast due to the prevailing westerly winds and the greater emission density in the east. The highest O_3 concentrations in the nation are found in the Los Angeles Basin of southern California, reflecting a combination of high emissions and poor ventilation.

We restrict our trend analysis to sites with at least one half-month of data (64 hourly data points) for each summer for a minimum of 12 years. Data for sequential years were sometimes available from sites a few kilometers apart, usually because the O_3 monitoring station had moved. In these cases, the O_3 data were merged. A total of 549 sites (Figure 1) were found suitable for trend analysis.

Our trend analysis focuses on the median and 90th percentile summer afternoon concentrations. Similar to [*Walker* 1985], ordinary least squares regression of O_3 concentrations versus year was used, and the slope of the regression line is reported as the trend, in

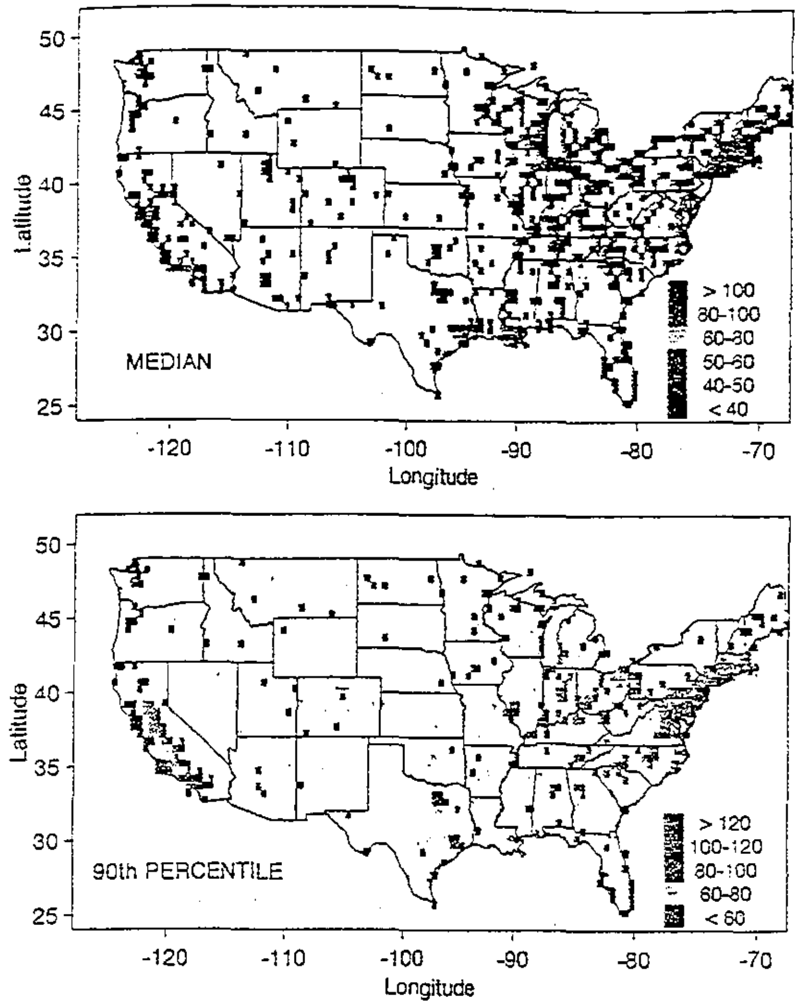

Plate 1. Summer afternoon O_3 concentrations (ppb) in surface air over the United States: (top) medians and (bottom) 90th percentiles. The maps were constructed from observations at 13-16 local time in June-August 1980-1995 for the sites of the EPA AIRS network. Data from individual sites were averaged over 50 x 50 km^2 grid squares.

both ppb per year and percent change per year. Percent changes are referenced to the mean value for the 1980-1995 period. A site was considered to possess a significant trend if the t-statistic for the slope of the regression line was equal to or above the 95% confidence limit. Figure 2 shows as an example the trends of 90^{th} percentile concentrations in Pasadena, California and Boston, Massachusetts.

The Boston data in Figure 2 display unusually high O_3 concentrations in 1988, when record-high temperatures and pollution were observed over much of the eastern United States [*Ludlum*, 1988]; this feature illustrates the importance of accounting for temperature variability when diagnosing O_3 trends. We referenced the daily O_3 concentrations measured at the individual AIRS sites to daily maximum surface temperatures from nearby National Climatic Data Center (NCDC) monitoring sites The NCDC sites were chosen for the length of their records and their proximity to AIRS sites (Figure 1). The O_3 concentrations at each AIRS site were thus sorted into 5 K temperature bins from 295 to 315 K. Median and 90^{th} percential concentrations were calculated for each bin that met the data density criteria discussed previously, and trends were analyzed within each bin. Only three bins contained enough data for useful analysis: 295-300 K, 300-305 K and 305-310 K. The temperature range 300-305 K included 75% of all AIRS trend sites (412/549) and will be the focus of our discussion.

The relationship between trends and local emissions was explored using two site classification systems. First, we used the EPA descriptions accompanying the AIRS database in which each site is labeled as "urban." "suburban," "rural," or "unknown." Second, we developed a more continuous classification of sites based on local emission data for anthropogenic NO_x and VOC from the National Acid Precipitation Assessment Program (NAPAP) [*EPA*, 1989]. For this purpose, we used the NAPAP summer weekday inventory including both area and point sources with 20x20 km^2 resolution. The two classification systems are compared in Figure 3. Sites designated as urban and suburban by the EPA tend to fall into the category of high local NO_x emissions (greater than 1 x 10^{12} molecules cm^{-2} s^{-1}), while rural sites are generally in the low NO_x emissions category (less than 5 x 10^{10} molecules cm^{-2} s^{-1}). Six sites considered rural by the EPA fall into the high NO_x category; four of these sites appear to be near power plants as indicated by their low VOC/NO_x ratios [*Sillman et al.*, 1990b]. The few sites classified by EPA as "unknown" tend to be in areas of low NO_x emissions.

3. Ozone Trends

The 1980-1995 trends in summer afternoon O_3 concentrations are shown in Plate 2 and 3 in terms of absolute magnitude (ppb per

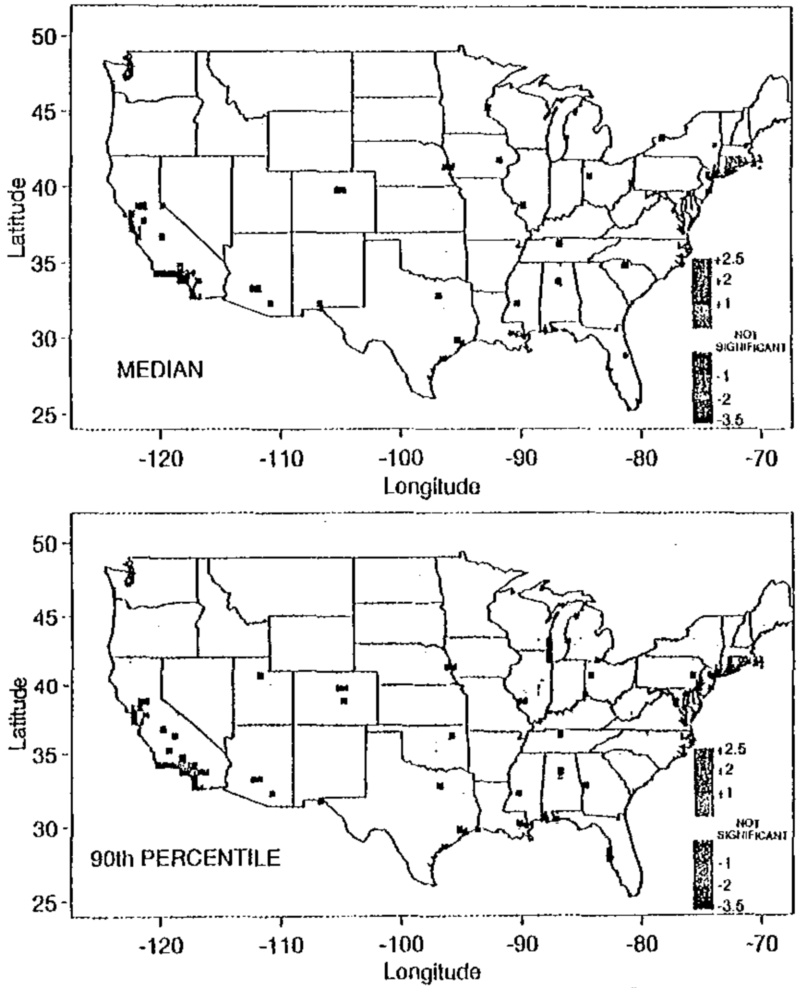

Plate 2. 1980-1995 trends (ppb per year) of median and 90th percentile O_3 concentrations for summer afternoon (13-16 local time) in surface air over the United States. The trends were computed for individual sites and results were averaged over 50 x 50 km^2 squares; sites with no significant trends were entered in the averages as having a trend of zero. The yellow shade indicates that no site in the 50x50 km^2 square had a significant trend.

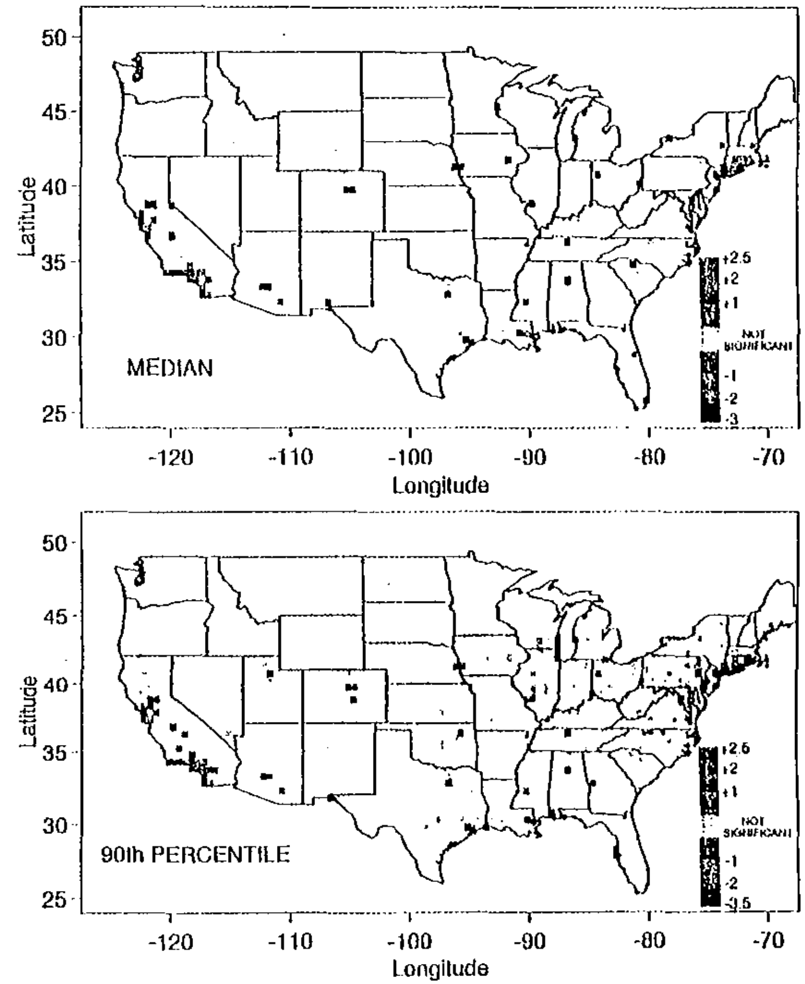

Plate 3. Same as Plate 2 but for relative trends in percent per year. The relative trends were referenced to the 1980-1995 mean of the corresponding quantity.

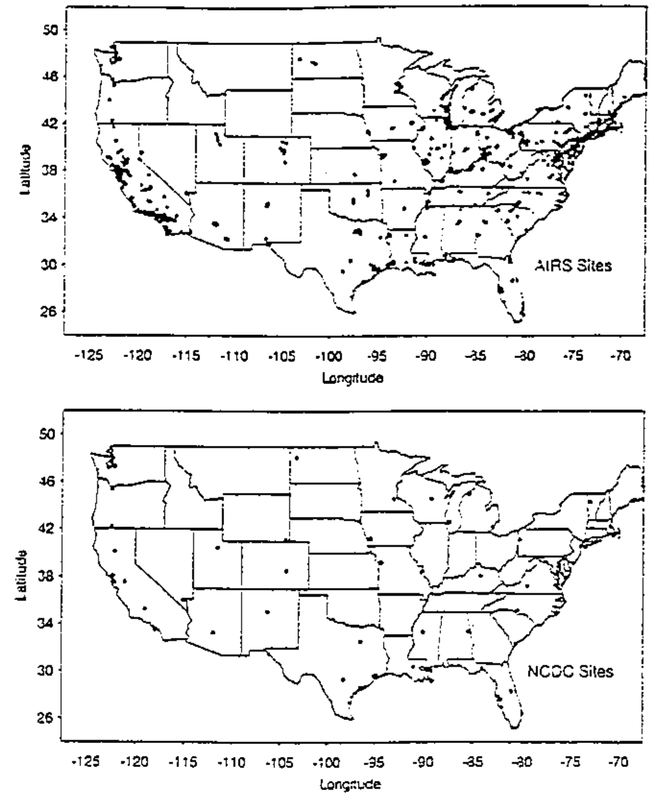

Figure 1. Ozone (EPA/AIRS) and temperature (NCDC) sites used in the trend analysis.

year) and relative magnitude (percent per year). Non-significant trends are shown in yellow. Table 2 gives the numbers and percentages of sites with significant trends. Increasing trends are detected at a small number of sites which are scattered nationwide: we do not find any worsening of air quality on a regional basis. Sites with decreasing trends tend to cluster around the large metropolitan areas. In both absolute and relative magnitude, the largest and most consistent decreasing trends are in the northeastern metropolitan corridor and in the Los Angeles Basin. More sites show significant downward trends in 90th percentile than in median concentrations, especially in the Chicago area. Half of all sites demonstrating

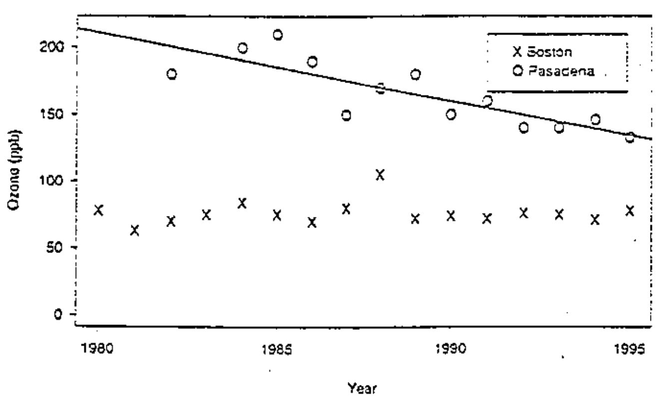

Figure 2. 1980-1995 trends of summer afternoon O_3 concentrations (90th percentile) in Pasadena, California, and Boston, Massachusetts. The Pasadena data show a significant decreasing trend (r^2 = 0.68; slope = -5.1 ppb yr^{-1}), while the Boston data do not (r^2 = 0.01).

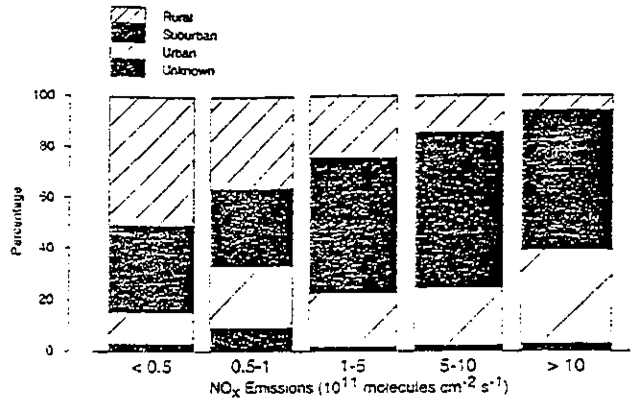

Figure 3. Relationship between the EPA/AIRS classification of sites ("urban," "suburban," "rural," "unknown") and the NAPAP NO_x emission inventory for 1985. Sites with NO_x emissions in a given range were sorted according to the EPA/AIRS classification.

improvements in 90th percentile O_3 levels are in the metropolitan areas of New York City (16 sites), Los Angeles (32 sites), and Chicago (5 sites). The remaining sites with significant decreasing trends are scattered throughout the nation and are generally urban or suburban.

Figure 4 provides a detailed view of the spatial distribution of trends in the metropolitan areas of New York City, Los Angeles, and Chicago. Similar patterns are found for the relative trends (not shown) in each of these three regions. In New York City, prevailing summer winds are from the southwest , and decreasing trends are observed for about 400 km downwind, spanning across Connecticut (Figure 4a). The largest trends are in coastal Connecticut, where median concentrations decreased by 1-1.5 ppb yr^{-1} over the 1980-1995 time period and 90th percentile concentrations decreased by 2-4 ppb yr^{-1}. In the Los Angeles Basin, the most pronounced decreases are 3 ppb yr^{-1} in median and 5 ppb yr^{-1} in 90th percentile concentrations (Figure 4b). Trends are strongest approximately 40 km inland and decline radially outward. The Chicago metropolitan area shows no significant trends in median O_3 concentrations and only scattered significant trends in 90th percentile concentrations (Figure 4c).

4. Segregation by Temperature

Trends in O_3 concentrations for days with maximum temperatures in the 300-305 K range are shown in Plate 4. Comparison with Plate 2 reveals many more decreasing trends in the data segregated by temperature. Table 2 gives the numbers of sites with significant trends in each temperature bin. We find that 17 and 32% of sites display downward trends in median and 90th percentile concentrations, respectively, as compared with 12 and 19% before segregation by temperature. Sites with downward trends in the temperature-segregated data are still clustered in the three metropolitan areas discussed above, but also extend to additional metropolitan areas including Detroit, Michigan; Cleveland, Ohio; Cincinnati, Ohio; Philadelphia, Pennsylvania; and Pittsburgh, Pennsylvania. The fraction of sites with upward trends is similar to the previous analysis. The improved detection of downward O_3 trends in the temperature-segregated data reflects interannual variability in temperature rather than any long-term warming trend: we find no significant warming for the 1980-1995 period at the NCDC sites used in our analysis.

Table 2. Numbers of Sites With Significant Trends

	Median[a]	Median 295–300 K	Median 300–305 K	Median 305-310 K	90th Percentile[a]	90th Percentile 295–300 K	90th Percentile 300–305 K	90th Percentile 305–310 K
Number of sites analyzed	549	187	412	198	549	187	412	198
Sites with increasing trends	20	2	11	10	7	0	5	2
	(4%)	(1%)	(3%)	(5%)	(1%)	(0%)	(1%)	(1%)
Sites with decreasing trends	67	45	72	26	106	81	131	42
	(12%)	(24%)	(17%)	(13%)	(19%)	(43%)	(32%)	(21%)

1980-1995 trends in medians and 90th percentiles of summer afternoon O_3 concentrations at the EPA AIRS sites across the United States. The percentages of sites with trends are shown in parentheses. The data were segregated by daily maximum temperature measured at nearby NCDC stations. Only sites with sufficient data over at least 12 years were analyzed (see text).

[a]Without segregation by temperature.

The bottom panels of Figure 4 display the spatial distribution of trends in the 300-305 K temperature range for the metropolitan areas of New York City, Los Angeles and Chicago. In the northeastern corridor, decreasing trends extend further southwest than in the previous analysis, incorporating Washington D. C. (Plate 4). In the Los Angeles Basin, the spatial distribution of trends is similar to that found in the previous analysis. It appears from Plate 4 that fewer sites exhibit downward trends south of the Los Angeles Basin

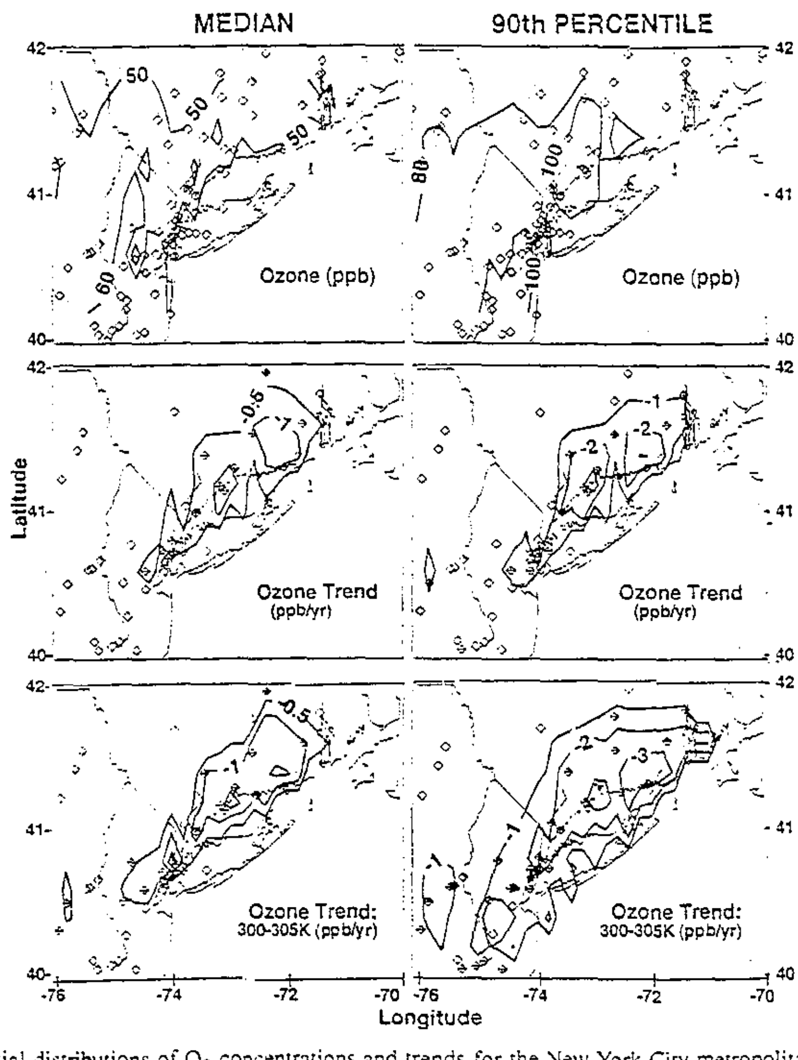

Figure 4a. Spatial distributions of O_3 concentrations and trends for the New York City metropolitan area in 1980-1995. The top panels show the median and 90th percentile summer afternoon concentrations, the middle panels show the corresponding trends, and the bottom panels show the trends in the temperature-segregated data (300-305 K temperature bin). Diamonds show the individual data sites: there are fewer trend sites than concentration sites because of the data density requirement. Crossed diamonds in the middle and lower panels indicate sites with significant decreasing trends.

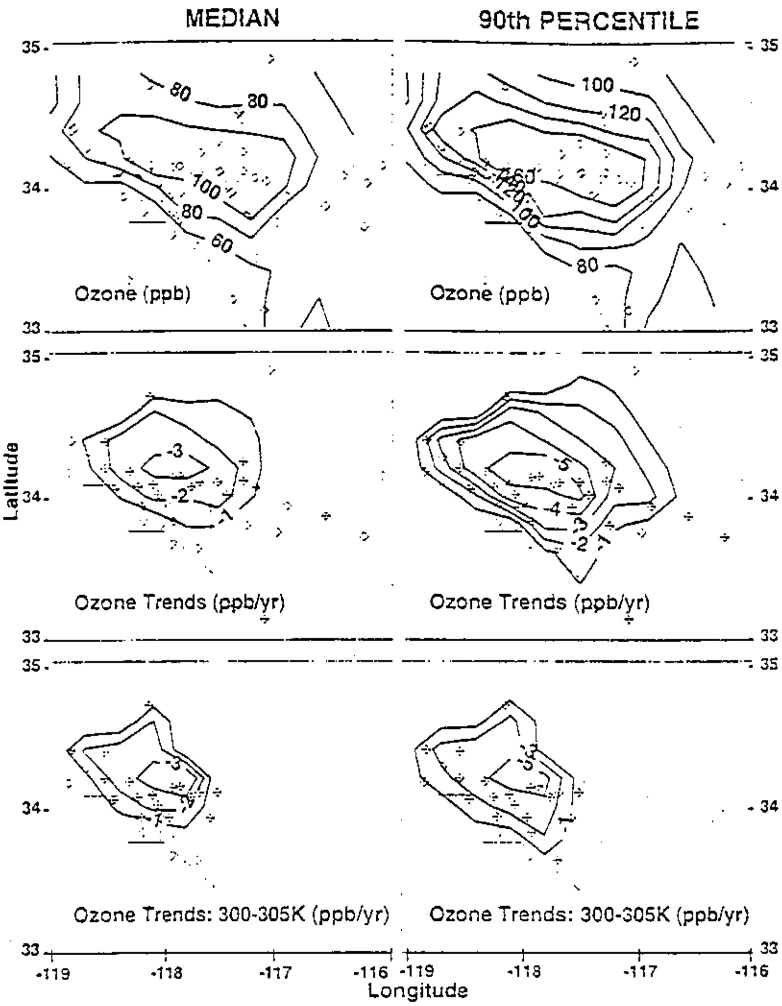

Figure 4b. Same as Figure 4a but for the Los Angeles Basin.

after segregation by temperature. Most summer days in this region have maximum temperatures in the range 305-310 K: decreasing trends are found within the 305-310 K bin, but the trends at the few sites with adequate data in the 300-305 K temperature bin are generally not significant. Many additional sites with decreasing trends emerge in the Chicago area (Figure 4c).

5. Relationship to Local Emissions

Figure 5 displays the percentage of sites with significant trends (positive or negative) sorted by local anthropogenic emissions of NO_x and VOC, local VOC/NO_x emission ratios, and EPA site classifications. The trends are for the data not segregated by temperature; results for the temperature-segregated data are similar. Increasing trends comprise a small percentage of most categories. Downward trends in the 90th percentile concentrations are more likely to be found at urban sites (high precursor emissions). Median concentrations are also more likely to show downward trends at urban than at rural sites according to the EPA classification; by the NO_x emission classification, however, approximately the same percentages of increasing and decreasing trends in median concentrations are found for all emission ranges.

Figure 5 indicates a correlation of trends with the VOC/NO_x emission ratio. This correlation could be taken to imply that O_3 air quality has improved most in areas where emissions are dominated by mobile sources (high ratios). However, we find that the correlation is determined mainly by the high VOC/NO_x emission ratios in the Los Angeles Basin. The large fraction of sites with decreasing trends in this region can be probably attributed to the more stringent emission controls than in the rest of the country, as discussed below, rather than to particularity in the VOC/NO_x emission ratio.

6. Discussion and Conclusions

Previous analyses of trends in summertime ozone are summarized in Table 1. Most of these studies used shorter records, and they used a variety of ozone statistics. The addition of a few years of data can lead to different trend results for such short time series, particularly if the analysis starts or ends near an anomalous year, such as 1988 which had record high ozone levels. The choice of ozone statistic (such as mean, daily maximum, seasonal extrema) and inclusion of explanatory variables in the trend model may also influence results. Our results are generally in accord with previous trend analyses, and differences are most likely attributable to the use of different time periods and choice of ozone statistic.

Our finding that no large region of the United States demonstrates significant increasing trends is consistent with the work of [*Lefohn and Shadwick* 1991] (Table 1), who found little evidence of significant trends in O_3 at 77 rural sites from 1979-1988. The few trends they did find were generally positive. As they noted, this

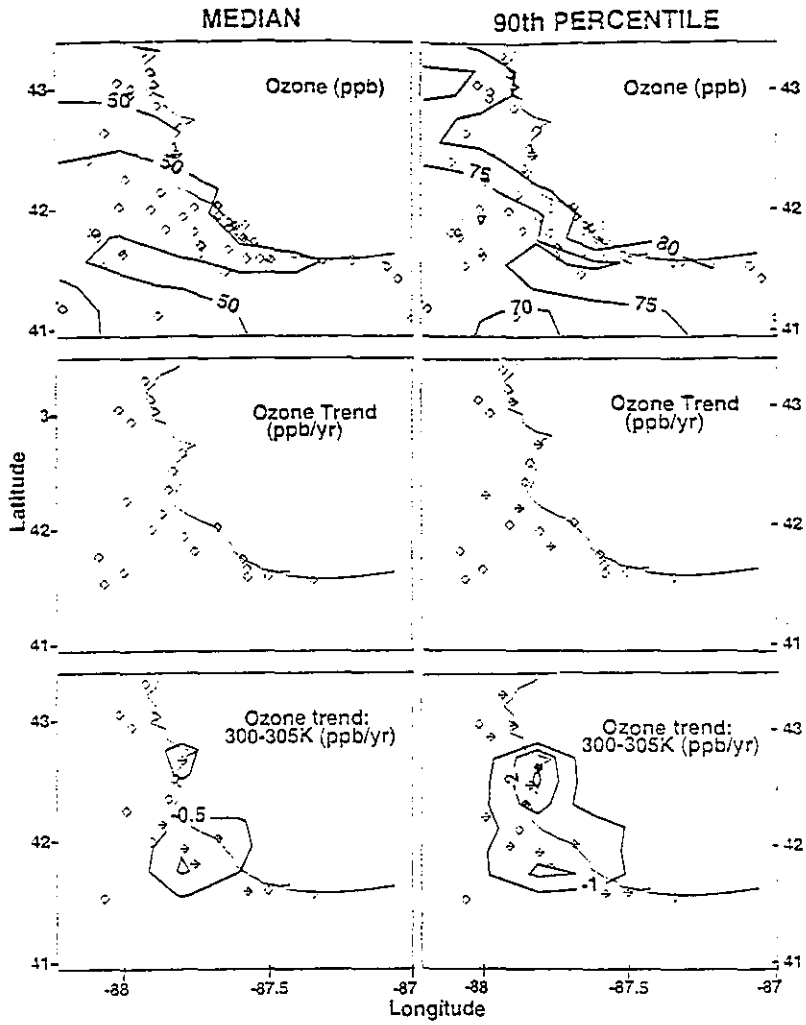

Figure 4c. Same as Figure 4a but for the Chicago area. No contours are shown in the middle panels because of the paucity of sites with significant trends.

result may have been influenced by the final year of their study, 1988, when O_3 concentrations were unusually high. Increases in ozone were found for only 2% of metropolitan areas for 1986-95 [*EPA*, 1996a]. We compared our results with trends in temperature-independent O_3 observed by *Rao et al.* [1995] in the eastern United States from 1983-1992 and found that we do not detect significant trends at roughly half of their sites. This discrepancy could reflect the 6 extra years of data in our analysis. Our findings compare well with the 1980-1992 trend analysis of *Zurbenko et al.* [1995] for the northeastern United States, where there are only 3 years of difference in the analysis periods.

Our conclusion that half the sites with decreasing trends in 90th percentile O_3 concentrations are in the metropolitan areas of New York City, Los Angeles, and Chicago is similar to results given by the Environmental Protection Agency [*EPA*, 1996a]. The EPA analysis gives trends in the second highest daily maximum O_3 value for 1986-95. They find decreases for 23% of 183 metropolitan statistical areas, and half the areas with decreases are in California, Chicago and environs, and the New York-New Jersey-Connecticut region. We find decreases in 90th percentile O_3 values for 19% of sites overall, again similar to *EPA* [1996a].

The downward trend of O_3 concentrations in the New York City metropolitan region has been reported previously by *Korsog and Wolff* [1991], *Rao et al.*, [1992, 1995], *Cox and Chu* [1993], *Rao and Zurbenko* [1994], *Zurbenko et al.* [1995], *EPA* [1996a] and *Flaum et al.* [1996]. A number of modeling studies have argued that the best strategy for controlling O_3 in this region is to reduce VOC emissions [*McKeen et al.*, 1991b; *Roselle et al.*, 1991; *Jacob et al.*, 1993; *Sillman*, 1993].

The Los Angeles Basin suffers from the most severe O_3 smog problem in the nation *EPA*, [1996a]. In an effort to ameliorate this situation, the state of California has implemented emission controls beyond the requirements for the rest of the nation. Total NO_x and VOC emission levels in the Los Angeles Basin in 1995 were 10 and 40% lower, respectively, than in 1980 (*B. Croes and D. Goodenow*, California Air Resources Board, personal communication, 1997). Our study indicates that these stringent regulations have met with success in reducing both median and extreme O_3 levels. *Walker* [1985], *Kuntasal and Chang* [1987], *Zeldin et al.* [1990] and *Cassmassi and Bassett* [1991] have previously reported downward trends in extreme concentrations of O_3 in the South Coast Air Basin.

The third largest metropolitan area of the nation (Chicago) shows significant downward trends for the 90th percentile O_3 concentrations but not for the medians. After segregation by temperature, more pronounced downward trends are found, although they

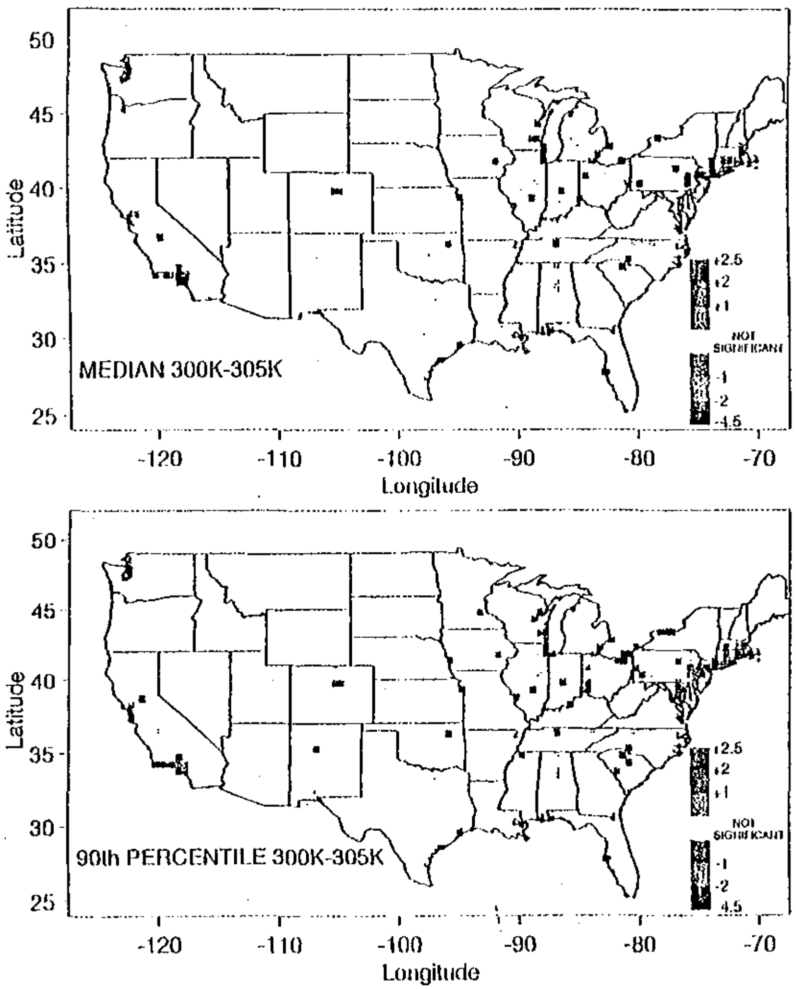

Plate 4. Same as Plate 2 but for days when the maximum temperature fell in the range 300-305 K.

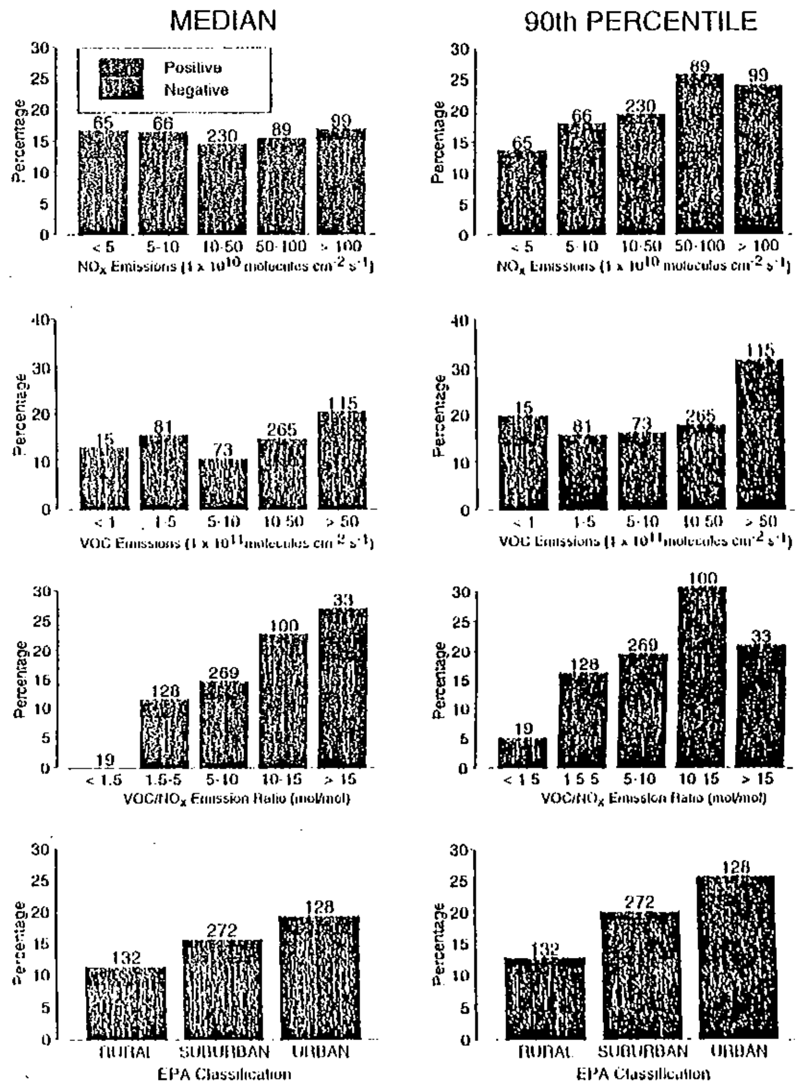

Figure 5. Percentage of sites with significant positive or negative trends, sorted by local NO_x and VOC anthropogenic emissions, local VOC/NO_x emission ratio, and EPA site classification. The total number of sites in each category is given at the top of each bar.

are still weaker than in New York City or Los Angeles. *Sillman* [1993] suggests that O_3 production in the Chicago urban plume should be VOC-sensitive.

In conclusion, we find decreasing trends in O_3 concentrations over the 1980-1995 period to be largely confined to the three largest metropolitan regions: New York City, Los Angeles, and Chicago. Within these regions, the trends are more pronounced for 90^{th} percentile concentrations (pollution episodes) than for the medians. National emission inventories indicate that VOC emissions declined by 12% over the 1980-1995 period while NO_x emissions remained constant. Although emission trends may vary regionally, the spatial distribution of O_3 trends in our analysis appears consistent with photochemical model calculations showing that O_3 production over the United States is NO_x-limited except in large urban plumes where it is partly VOC-limited [*Trainer et al.*, 1987; *Milford et al.*, 1989; *McKeen et al.*, 1991a,b; *Roselle et al.*, 1991; *Jacob et al.*, 1993]. VOC-limited conditions in urban plumes persist longer under conditions of low dispersion associated with pollution episodes in large metropolitan areas [*Jacob et al.*, 1993; *Sillman and Samson*, 1995]. Abatement of O_3 pollution elsewhere in the country appears contingent upon a reduction of NO_x emissions.

Acknowledgements. We thank Wendy McDougall for supplying the EPA AIRS data and Rebecca Taylor for obtaining the NCDC data. This work was supported by Harvard undergraduate research fellowships to A.M. Fiore (Harvard College Research Program, Harvard Dean's Summer Research Award), by an undergraduate research fellowship to J.H. Yin from the National Institute for Global Environmental Change, and by the National Science Foundation (NSF-ATM-93-20778 and NSF-ATM-96-12282).

References

Calvert, J.G., J.B. Heywood, R.F. Sawyer, and J.H. Seinfeld, Achieving acceptable air quality: Some reflections on controlling vehicle emissions, *Science, 261,* 37-45, 1993.

Cassmassi, J.C., and M. Bassett, Air quality trends in the South Coast Air Basin, in *Southern California Air Quality Study Data Analysis: Proceedings of an International Specialty Conference*, pp. 1-6, Air & Waste Manage. Assoc., Pittsburgh, Pa., 1991.

Chameides, W.L., et al., Ozone precursor relationships in the ambient atmosphere, *J. Geophys. Res., 97,* 6037-6055, 1992.

Chameides, W.L., et al., Ozone pollution in the rural United States and the new NAAQS, *Science, 276,* 916, 1997.

Chock, D.P., The need for a more robust ozone air quality standard, *J. Air Pollut. Control Assoc., 39,* 1063-1072, 1989.

Clark, T.L., and T.R. Karl, Application of prognostic meteorological variables to forecasts of daily maximum one hour ozone concentrations in the northeastern United States, *J. Appl. Meteorol., 21,* 1662-1671, 1982.

Cox, W.M., and S.H. Chu, Meteorologically adjusted ozone trends in urban areas: A probabilistic approach, *Atmos. Environ., 27,* 425-434, 1993.

Environmental Protection Agency (EPA), The 1985 NAPAP emission inventory (version 2): Development of the annual data and modeler's tapes, *EPA Rep. EPA-600/7-89-012a*, Research Triangle Park, N.C., 1989.

EPA, National air quality and emissions trends report: 1995, *EPA Rep. EPA-454/R-96-005*, Research Triangle Park, N.C., 1996a.

EPA, National air pollutant emission trends, 1900-1995, *EPA Rep. EPA-454/R-96-007*, Research Triangle Park, N.C., 1996b.

Flaum, J.B., S.T. Rao, and I.G. Zurbenko, Moderating the influence of meteorological conditions on ambient ozone concentrations, *J. Air Waste Manage. Assoc., 46,* 35-46, 1996.

Jacob, D.J., J.A. Logan, G.M. Gardner, R.M. Yevich, C.M. Spivakovsky, S.C. Wofsy, S. Sillman, and M.J. Prather, Factors regulating ozone over the United States and its export to the global atmosphere, *J. Geophys. Res., 98,* 14,817-14,826, 1993.

Korsog, P.E., and G.T. Wolff, An examination of urban ozone trends in the northeastern U.S. (1973-1983) using a robust statistical method, *Atmos. Environ., 25,* 47-57, 1991.

Kuntasal, G., and T.Y. Chang, Trends and relationships of O_3, NO_x and HC in the South Coast Air Basin of California, *J. Air Pollut. Control Assoc., 37,* 1158-1163, 1987.

Lefohn, A.S., and D.S. Shadwick, Ozone, sulfur dioxide, and nitrogen dioxide trends at rural sites located in the United States, *Atmos. Environ., 25,* 491-501, 1991.

Lippmann, M., Health effects of tropospheric ozone, *Environ. Sci. Technol., 25,* 1954-1962, 1991.

Logan, J. A., Ozone in rural areas of the United States, *J. Geophys. Res., 94,* 8511-8532, 1989.

Ludlum, D.M., Weatherwatch: July, August 1988, *Weatherwise, 41,* 300-309, 1988.

McKeen, S.A., E.-Y. Hsie, and S.C. Liu, A study of the dependence of rural ozone on ozone precursors in the eastern United States, *J. Geophys. Res., 96,* 15,377-15,394, 1991a.

McKeen, S.A., E.-Y. Hsie, M. Trainer, R. Tallamraju, and S.C. Liu, A regional model study of the ozone budget in the eastern United States, *J. Geophys. Res., 96,* 10,809-10,845, 1991b.

Milford, J.B., A.G. Russell, and G.J. McRae, A new approach to photochemical pollution control: Implications of spatial patterns in pollutant responses to reductions in nitrogen oxides and reactive organic gas emissions, *Environ. Sci. Technol., 23,* 1290-1301, 1989.

National Research Council (NRC), *Rethinking the Ozone Problem in Urban and Regional Air Pollution*, Nat. Acad. Press, Washington, D. C., 1991.

Prinz, B., Ozone effects on vegetation, in *Tropospheric Ozone*, edited by I.S.A. Isaksen, pp. 161-164, D. Reidel, Norwell, Mass., 1988

Rao, S.T., and I.G. Zurbenko, Detecting and tracking changes in ozone air quality, *J. Air Waste Manage. Assoc., 44,* 1089-1092, 1994.

Rao, S.T., G. Sistla, and R. Henry, Statistical analysis of trends in urban ozone air quality, *J. Air Waste Manage. Assoc., 42,* 1204-1211, 1992.

Rao, S.T., E. Zalewsky, and I.G. Zurbenko, Determining trends and spatial variations in ozone air quality, *J. Air Waste Manage. Assoc., 45,* 57-61, 1995.

Roselle, S. J., T.E. Pierce, and K.L. Schere, The sensitivity of regional ozone modeling to biogenic hydrocarbons, *J. Geophys. Res., 96,* 7371-7394, 1991.

Sillman, S., Tropospheric ozone: The debate over control strategies, *Annu. Rev. Energy Environ., 18,* 31-56, 1993.

Sillman, S., and P. J. Samson, Impact of temperature on oxidant photochemistry in urban, polluted rural and remote environments, *J. Geophys. Res., 100,* 11,497-11,508, 1995.

Sillman, S., J.A. Logan, and S.C. Wofsy, The sensitivity of ozone to nitrogen oxides and hydrocarbons in regional ozone episodes, *J. Geophys. Res., 95,* 1837-1851, 1990a.

Sillman, S., J.A. Logan, and S.C. Wofsy, A regional scale model for ozone in the United States with subgrid representation of urban and power plant plumes, *J. Geophys. Res., 95,* 5731-5748, 1990b.

Tietenberg, T., *Environmental and Natural Resource Economics*, Harper-Collins College, New York, 1996.

Trainer, M., E.J. Williams, D.D. Parrish, M.P. Buhr, E.J. Allwine, H.H. Westberg, F.C. Fehsenfeld, and S.C. Liu, Models and observations of the impact of natural hydrocarbons on rural ozone, *Nature, 329,* 705-707, 1987.

Walker, H.M., Ten-year ozone trends in California and Texas, *J. Air Pollut. Control Assoc., 35,* 903-912, 1985.

Wolff, G.T., and P.J. Lioy, An empirical model for forecasting maximum daily ozone levels in the northeastern U.S., *J. Air Pollut. Control Assoc., 28,* 1034-1038, 1978.

Zeldin, M.D., J.C. Cassmassi, and M. Hoggan, Ozone trends in the South Coast Air Basin: An update, in *Tropospheric Ozone and the Environment: Papers from an International Conference*, edited by R.L. Berglund, D.R. Lawson, and D.J. McKee, pp. 1-12, Air & Waste Manage. Assoc., Pittsburgh, Pa, 1990.

Zurbenko, I.G., S.T. Rao, and R.F. Henry, Mapping ozone in the eastern United States, *Environ. Manager, 1,* 24-30, 1995.

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(Received July 7, 1997; revised October 3, 1997; accepted October 13, 1997.)