

Draft

**ANALYSIS OF
FACTORS INFLUENCING OZONE
TRENDS IN THE TYLER/LONGVIEW/MARSHALL
NEAR NONATTAINMENT AREA**

Prepared for

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1. INTRODUCTION

This report describes results of a review and analysis of air quality and meteorological conditions and trends and ozone precursor emission trends in Northeast Texas. The Texas Natural Resource Conservation Commission (TNRCC) operates three Continuous Air Monitoring Stations (CAMS) in the Tyler/Longview/Marshall (TLM) area of East Texas. These stations monitor compliance with the National Ambient Air Quality Standard (NAAQS) for ozone. In recent years ozone levels measured at the Gregg County Airport (Longview) have exceeded the 1-hour ozone NAAQS. In 1996 the TLM area became a Flexible Attainment Region (FAR) and a mechanism for developing strategies to attain the 1-hour ozone standard was implemented under a Memorandum of Agreement (Flexible Attainment Region Memorandum of Agreement, September 16, 1996).

The TLM area has received funding from the Texas legislature to address ozone air quality issues. These resources have funded studies through the East Texas Council of Governments (ETCOG) under the technical and policy direction of the North East Texas Air Care (NETAC) organization. In 1999, ENVIRON completed an ozone modeling study for two 1-hour ozone episodes that included future year modeling for 2007 and the evaluation of future year emission reduction strategies (Yarwood et al., 1999). In May 2002, the TNRCC submitted a State Implementation Plans (SIP) for Northeast Texas that demonstrated attainment of the 1-hour ozone standard by 2007 (TNRCC, 2002). In addition to these analyses, TNRCC has been studying air quality conditions and trends in the region (Dave Sullivan, TNRCC, personal communication, 2002).

In 1997 the U.S. Environmental Protection Agency (EPA) promulgated a new 8-hour NAAQS for ozone that is potentially much more stringent than the existing 1-hour standard. The 8-hour ozone NAAQS was challenged in court and was eventually upheld in 2002 by the U.S. Supreme Court. However, the Court required that the EPA revise its implementation policy. Details of the implementation are still evolving, but it is now clear that designated regions will have to prepare and submit SIPs for 8-hour ozone. TLM is one of the five "near" nonattainment areas (NNAs) in Texas that have been preparing technical studies to support development of an 8-hour ozone SIP should this become necessary.

The objective of this study is to support SIP preparation activities for TLM by examining the historical record of ozone, emissions and meteorological data in the region to explain the changing ozone levels at the Gregg County airport (Longview) monitor. This location has historically recorded the highest ozone levels in the NNA. The frequency of high 1-hour ozone events at Longview has varied widely from year to year with some recent years having several exceedances of the 1-hour ozone standard and other years having none. Much of this variability is attributable to meteorology (e.g., temperature, wind speed, wind direction) but other factors may be contributing. In this study we review the historical record of ozone, emissions and meteorology in the TLM NNA from 1980 – 2001, looking for relationships that explain the changes in the frequency of high ozone levels over time. Ozone trends in TLM are also compared and contrasted with trends in two nearby population centers (Dallas and Shreveport).

2. DATA COLLECTION

AIR QUALITY AND METEOROLOGICAL DATA

As noted in Section 1, the Texas Natural Resource Conservation Commission (TNRCC) has operated three continuous air monitoring stations (CAMS) in the study region over the past few years (see map in **Figure 2-1**). Historically, the highest ozone concentrations have been recorded at the Longview monitor (CAMS-19) which is located at the Gregg County airport. Ozone measurements have been collected at this site since the 1970s. An SO₂ monitor was added at this location in 1999. Ozone monitoring commenced in 1995 at Tyler Airport (CAMS-86) and in 1998 at the Cypress River Airport (CAMS-50). Note that the Cypress River monitor was discontinued in March 2001 and a new site located across the county line in Harrison County (Karnack, CAMS-85) began operating in September 2001. The Tyler monitor was relocated due to airport construction and assigned a new number (CAMS-82) in 2000.

A data base of daily data, including daily maximum 1-hour and daily maximum 8-hour average ozone concentrations, number of valid hours of ozone data, and daily maximum, minimum, and average temperatures were provided for this study by David Sullivan of the TNRCC. Hourly ozone, SO₂, wind speed and direction data for selected years were downloaded from the TNRCC web site.

Results of trends and other analyses of the air quality and meteorological data are presented in Section 3.

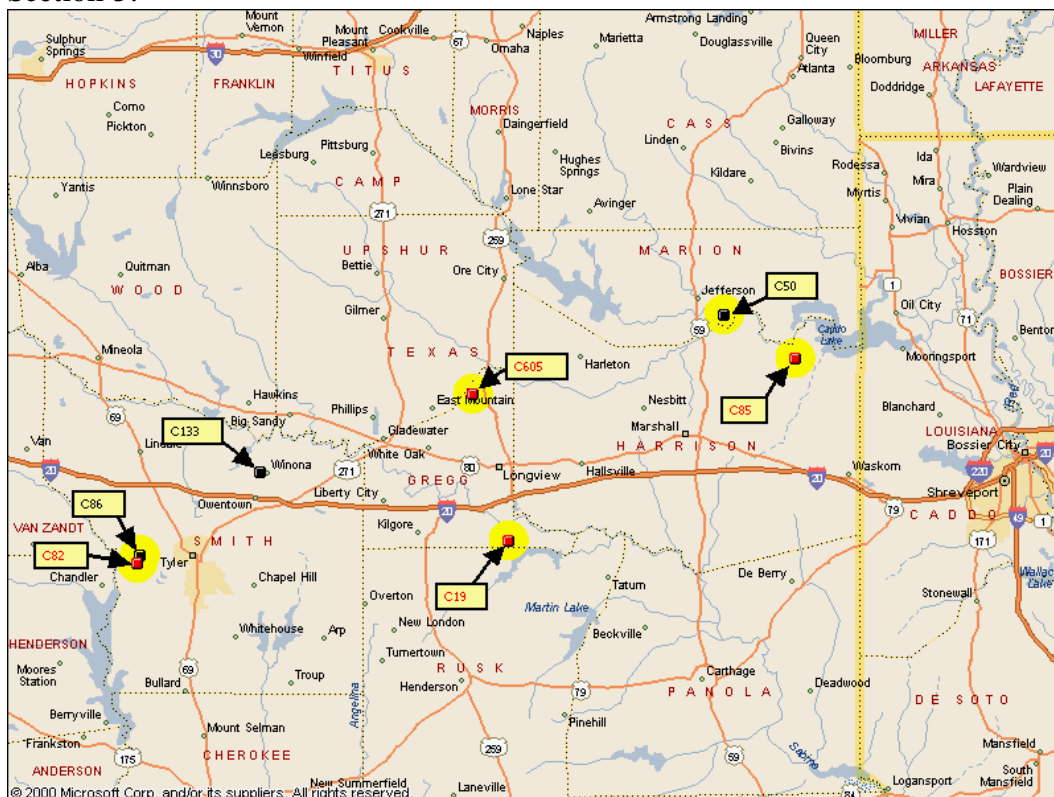


Figure 2-1. Map of Northeast Texas showing locations of TNRCC air quality monitors (source: www.tnrcc.state.tx.us).

EMISSIONS DATA

Development of a comprehensive, retrospective emissions inventory for the 1980 – 2001 study period is impractical because of large changes in inventory methods and standards over this timeframe. However, we were able to assemble a working inventory of NO_x and VOC emissions in the region using data from various sources along with some reasonable assumptions about source output during years for which actual data were unavailable.

Emissions estimates for area, on-road mobile, non-road mobile, and most of the major point sources in the five county study region (consisting of Gregg, Harrison, Rusk, Smith, and Upshur counties) were obtained from a variety of sources. **Figure 2-2** shows the locations of the major stationary sources relative to the ambient air quality monitoring sites. Annual average daily nitrogen oxides (NO_x) emissions for all of the large utility boilers in the region (Knox Lee, Pirkey, and Martin Lake) were assembled from information provided by utility plant operators and EPA's Acid Rain Program. In general, emission estimates for these sources became more accurate after introduction of continuous emission monitors in 1995 than was the case in earlier years. Annual emissions estimates for Pirkey were provided for 1990 – 2001; emissions for 1985 (the year the plant began operations) were computed by calculating an emission factor based on reported 1990 emissions and 1990 heat input obtained from EPA's Acid Rain database and applying it to the 1985 heat input value from the Acid Rain database. Emissions for 1986 – 1989 were assumed to be equal to the 1990 level. Annual emissions data for Knox Lee were available for 1994 – 2001; emissions in prior years were set equal to the 1994 – 2001 average. Annual emissions data for Martin Lake were available from 1982 – 2001; emissions for 1981 were assumed equal to 1982.

Emissions of NO_x and volatile organic compounds (VOC) from Texas Eastman (a large chemical plant located roughly 4-km to the north-northeast of the Longview monitor) for the period 1988-2001 were provided by Sharon Wellman of Texas Eastman. Emissions from this facility prior to 1988 were simply assumed to have remained constant at the 1988 level.

Daily emissions were adjusted to ozone season daily averages for sources for which seasonal adjustment factors were available. Emissions from other stationary sources in the five county region were available only for 1996 – 1999. During this period, these sources accounted for an average of 8 % of inventory total NO_x emissions (22 tons/day). We assumed that emissions from these miscellaneous stationary sources remained constant at this level during all other years of the study period.

Non-road mobile and area source emissions for 1990 – 2001 for the five county region were obtained from an inventory development effort completed last year (ENVIRON, et al., 2001). We did not make any inferences about emissions from these source categories prior to 1990 since their impact on total emissions is not insignificant and additional information on trends during the earlier years was not readily available.

On-road mobile source emissions in the five county study region for 1990 – 2001 for an average summer weekday were estimated using the MOBILE 6 emission factor model with model inputs applicable to the study region as described in (ENVIRON, et al., 2001) and county total VMT data for 1981 – 2001 provided by the Texas Department of Transportation (TXDOT). This work was extended back to 1981 using TXDOT VMT data for the earlier years. MOBILE inputs for the earlier years were developed on the basis of the 1990 – 2001

inputs with appropriate adjustments for changes in Reid vapor pressure (RVP). The 1989 RVP value in the study region was 9.0 (this was lowered to 7.8 in 2000). Region-specific RVP data was not available prior to 1989 (i.e., prior to institution of a specific regulatory limit) so we set the RVP equal to the national average value of 10.9. On-road emission trends are illustrated in **Figure 2-3**. Significant VOC reductions occurred due to replacement of older vehicles with newer models with lower tailpipe and evaporative emissions and due to the introduction of lower RVP gasoline. NO_x reductions over this period have been much smaller.

Biogenic NO_x and VOC emissions were not included in the inventory development effort since it is reasonable to assume that there were no significant trends in biogenics during the 21 year period. Of course, emissions from biogenics will change significantly from day to day and, to a lesser extent from year to year due to changes in meteorological conditions. During the summer months, biogenics are the dominant VOC source in the region, accounting for about 900 Tons/Day (Appendix A) or about 85% of total VOC emissions (1996 average ozone season day).

Summer average day emissions for sources described above are listed in **Table 2-1**. A summary of VOC and NO_x emissions by major source category for 1999 (the most recent year for which emissions data for the smaller point sources were readily available) is presented in **Figure 2-4**. Point sources contribute more than half of total NO_x with the remainder approximately equally divided between area/non-road and on-road mobile sources. Area and non-road sources contribute half of the total VOC with the rest approximately equally divided between on-road mobile and point sources. Summaries of emission trends are presented in Section 3.

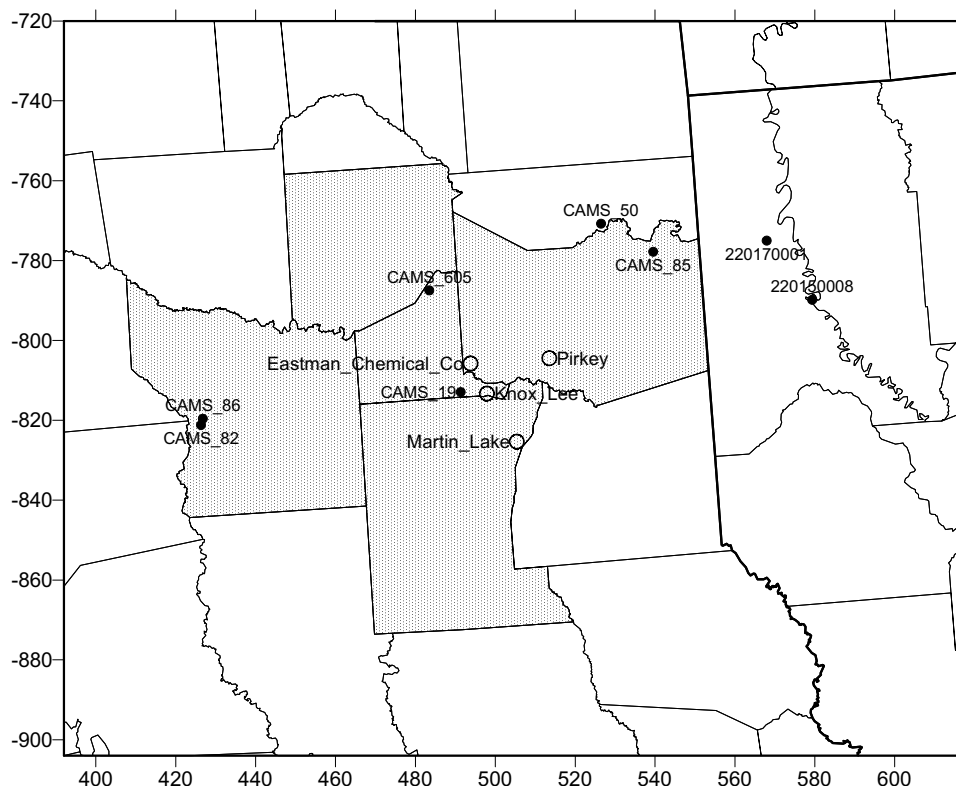


Figure 2-2. Northeast Texas air quality monitoring sites (filled circles) and major point sources (open circles). Counties in the NNA are shaded.

Table 2-1. Summer season emissions in Tyler-Marshall-Longview NNA (average tons/day).¹

Year	On-Road Mobile		Non-Road and Area ²		Eastman Chemical ³		Pirkey ⁴	Knox Lee ⁵	Martin Lake ⁶	Other Point Sources ⁷
	VOC	NOx	NOx	VOC	VOC	NOx	NOx	NOx	NOx	NOx
1981	114	76			22	36	0	4	128	7
1982	114	77			22	36	0	4	128	7
1983	116	78			22	36	0	4	129	7
1984	116	78			22	36	0	4	128	7
1985	113	77			22	36	18	4	109	7
1986	106	73			22	36	18	4	95	7
1987	100	71			22	36	18	4	89	7
1988	95	69			22	36	18	4	94	7
1989	73	67			20	31	18	4	97	7
1990	69	64	81	97	18	27	18	4	89	7
1991	60	58	78	92	15	24	17	4	91	7
1992	55	56	80	94	12	21	21	4	91	7
1993	53	56	83	95	12	19	23	4	83	7
1994	52	58	78	91	11	17	23	4	88	7
1995	49	58	74	89	11	16	20	2	88	7
1996	45	57	73	90	11	16	31	3	82	6
1997	43	56	71	86	10	16	27	4	90	12
1998	42	58	69	84	10	16	29	4	85	6
1999	40	56	67	80	10	16	28	5	77	7
2000	35	54	64	74	10	15	29	5	75	7
2001	31 ⁸	52 ⁸	62	74	9	12	23	4	60	7

¹ Where seasonal data were not available (e.g., for certain point sources), annual average day emissions are reported.

² From ENVIRON (2001); estimates for years prior to 1990 are unavailable.

³ Estimates for 1988-2001 provided by Sharon Wellman of Eastman Chemical; estimates for 1981-1987 set equal to 1988 levels.

⁴ Estimates for 1990 – 2001 provided by Howard Ground, AEP with values prior to 1993 adjusted to reflect emission factor calculated from 1994 CEM data; 1985 emissions estimated from 1985 heat input data reported to EPA's Acid Rain Program and adjusted 1990 emission factor; estimates for 1986-1989 based on 1990 level; this plant began operation in 1985.

⁵ Estimates provided by Howard Ground, AEP; values prior to 1994 based on 1994 – 2001 average.

⁶ Estimates for 1995-2001 based on CEM data; emission for prior years estimated from NOx emission factors and heat input data. All data for this plant provided by David Duncan, Texas Utilities.

⁷ Estimates for 1996-1999 from TNRCC PSDB data; values for other years set equal to 1996-1999 average.

⁸ Extrapolated from 1998-2000 trend.

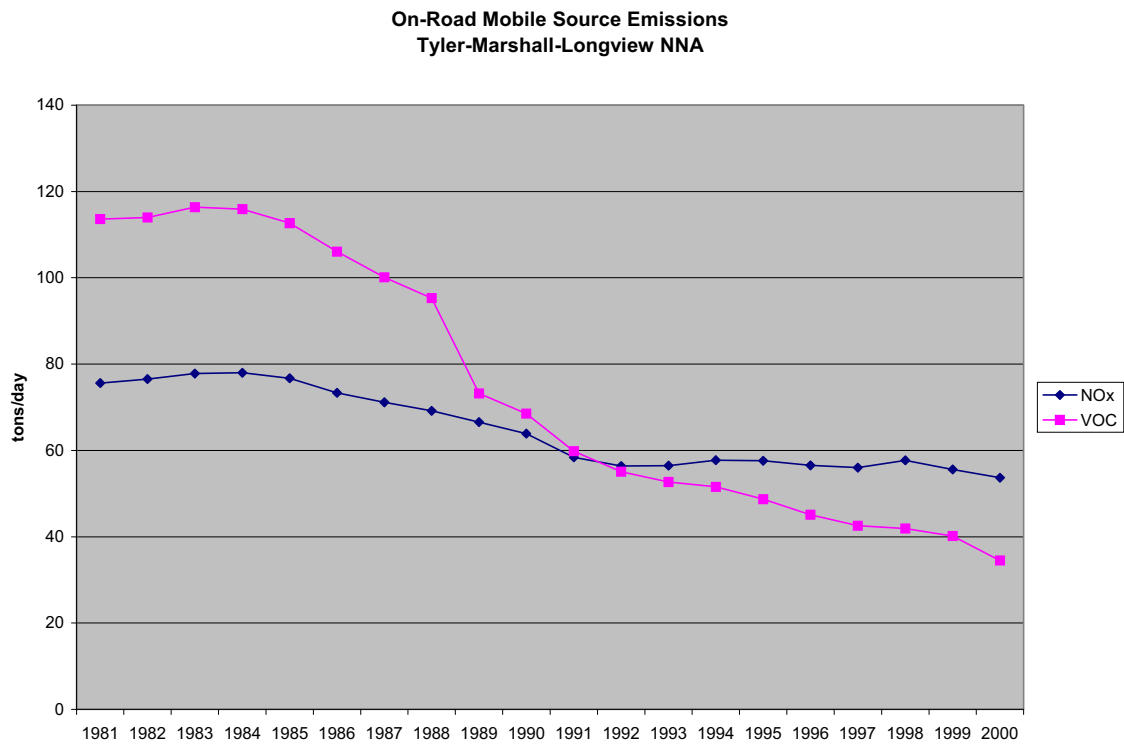


Figure 2-3. On-road mobile source emission trends in the Tyler-Marshall-Longview NNA (average summer day).

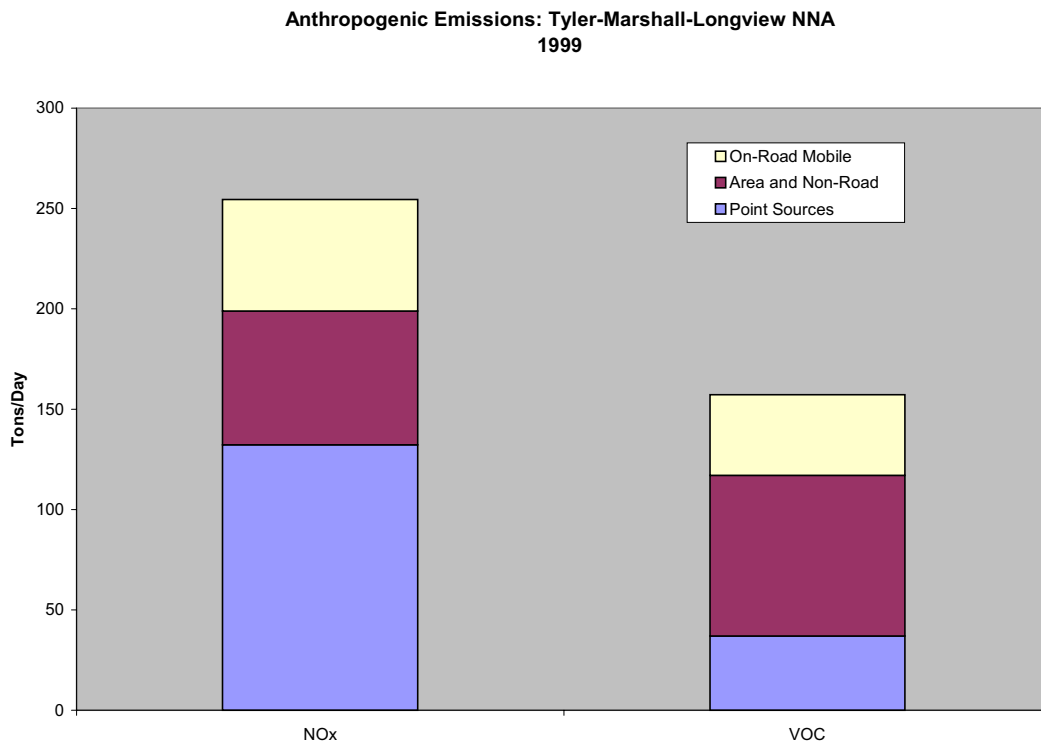


Figure 2-4. Summary of 1999 average summer day NOx and VOC emissions by major source categories in the Tyler-Marshall-Longview NNA.

3. RESULTS AND DISCUSSION

Analyses of trends in emissions of ozone precursors in the Tyler-Marshall-Longview NNA are described in the following subsection. This is followed by an analysis of trends in ambient ozone, relationships between ozone and meteorological conditions and between ozone and SO₂.

EMISSION TRENDS

A rough estimate of anthropogenic emission trends in the five county study area was assembled from a variety of sources as described in Section 2 above. NO_x emission trends for the study region are summarized in **Figure 3-1**. This figure does not show area and non-road emissions prior to 1990 because there is too much uncertainty to back-cast these emissions, as discussed in Section 2. Between 1981 and 2001 NO_x emissions from on-road mobile and point sources combined are estimated to have decreased nearly 40%. On-road mobile source NO_x emissions decreased 31% during this period. Non-road and area source NO_x emissions during 1990 – 2001 are not estimated to have appreciably altered the overall downward trend. Total NO_x emissions from all inventoried sources are estimated to have decreased 27% between 1990 and 2001.

Figure 3-2 displays available data on VOC emission trends. Data for Texas Eastman (the largest anthropogenic VOC point source in the area) are available for 1988 – 2001; data for non-road mobile and area sources are only available for 1990 - 2001. Mobile source data were estimated back to 1980 as described in Section 2. VOC emissions from on-road mobile sources and Eastman combined are estimated to have declined 66% from 1988 to 2001; during 1990 – 2001, total VOC from all anthropogenic sources (excluding stationary sources other than Eastman) are estimated to have declined 38%. Similar trends cannot be computed for the full 21 year period due to the lack of area and non-road emissions data in the 1980s, but it is interesting to note that the decline noted above in VOCs from on-road mobile and Eastman between 1988 and 2001 (66%) is much greater than the decline in NO_x from these sources over the same period (37%); adding the estimated point source NO_x into this trend does not change the overall NO_x reduction factor appreciably (40% decline). Thus, as far as anthropogenic emissions go, the VOC/NO_x ratio has decreased appreciably during this period. We note, however, that on a regional scale this change occurred against a constant background of biogenic VOCs which are estimated to account for 85% of total VOC emissions. The contribution of anthropogenic emissions to total VOC emissions has decreased over the years as anthropogenic VOC emissions have declined.

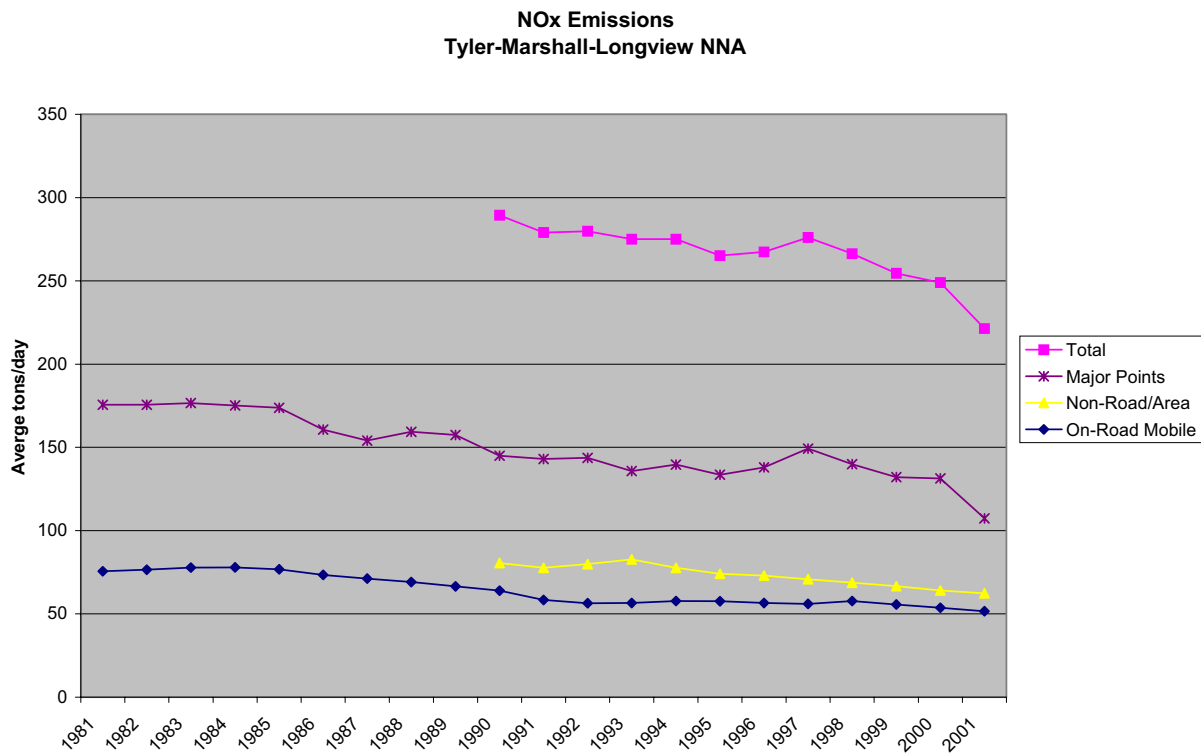


Figure 3-1. NOx emission trends from 1981-2001 in the Tyler-Marshall-Longview NNA (average summer day).

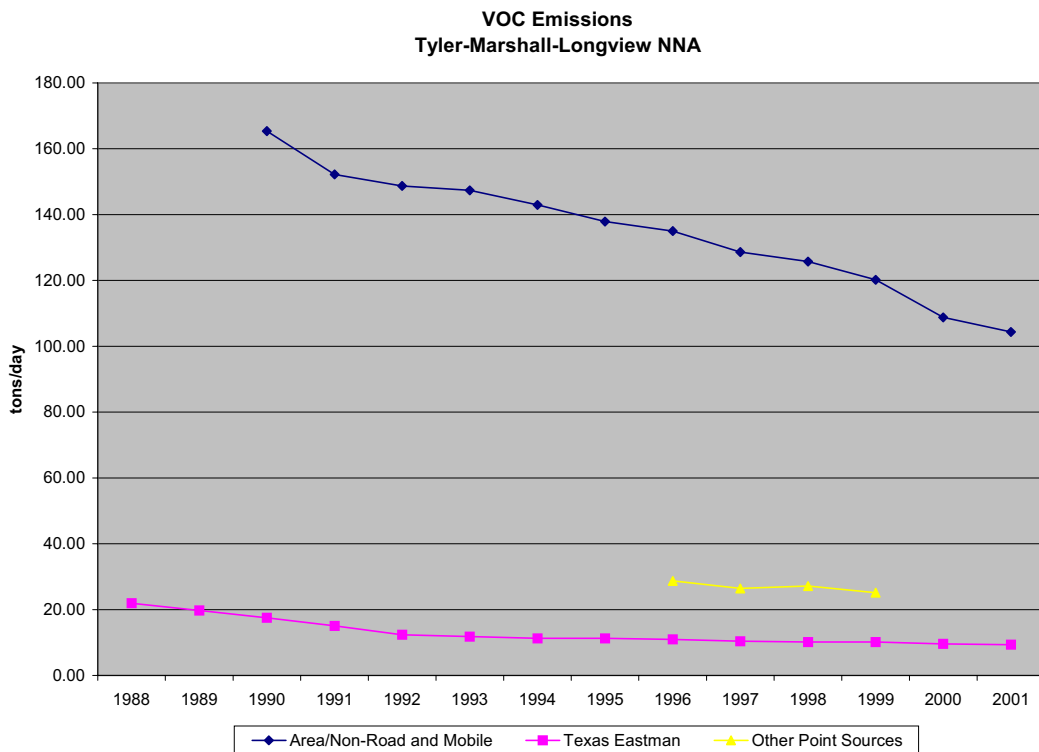


Figure 3-2. Anthropogenic VOC emission trends from 1988-2001 in the Tyler-Marshall-Longview NNA (average summer day).

OZONE TRENDS

Our analysis of ozone trends focussed primarily on data collected at the Longview (CAMS-19) monitoring site since this is the only long-term site in the TLM NNA. Other sites in the area came on-line starting in 1995. EPA's National Ambient Air Quality Standard for ozone includes both a 1-hour average standard and an 8-hour average standard. The 1-hour standard limits the frequency with which the daily maximum 1-hour average concentration can exceed 0.12 ppm to once per year (averaged over three years) while the 8-hour standard sets a maximum level (0.08 ppm) for the annual fourth highest daily maximum 8-hour average concentration.¹ The 1-hour standard is violated if the fourth highest concentration in a period of three consecutive years exceeds 0.12 ppm. Although a single year of data is not considered sufficient to demonstrate attainment, the value of the second highest daily maximum 1-hour concentration in a year is frequently used as an informal indicator of attainment/nonattainment status. In the following discussion, we refer to this as the annual 1-hour design value. The 8-hour standard is violated if the annual fourth highest daily maximum 8-hour average concentration averaged over three consecutive years exceeds 0.08 ppm. Again, although a single year of data is not considered sufficient to demonstrate attainment, the fourth highest value in a given year is frequently used as an informal indicator of attainment/nonattainment status. Consequently, in the following discussion we refer to this statistic as the annual 8-hour design value.

Eight-Hour Design Value Trends

Figure 3-3 shows the 1980 – 2001 trend in the annual fourth highest daily maximum 8-hour average ozone concentrations at the Longview Airport monitor (CAMS 19; AIRS ID 481830001). Years in which the monitoring data do not meet EPA's 75% completeness criterion (40 CFR 50, Appendix I) are plotted as squares. The trend in annual 8-hour design values shows a decline in ozone concentration through 1993 or 1994 followed by a period of widely fluctuating values. Design values in 1998 and 1999 were the highest of any during this 22 year period; the lowest values occurred in 1993 and 1994. Visual inspection of **Figure 3-3** suggests two very different trend periods at this monitoring site. For the 15-year period from 1980 until 1994, design values experienced a gradual but relatively steady decline. Beginning in 1995, however, the design values fluctuate widely, reaching values in four of the seven years which exceeded any observed during the entire prior period. For 1980 – 1993 a least squares regression gives a slope of -0.87 ppb/year with a 95% confidence interval of $(-1.3$ ppb/year to 0.43 ppb/year), the slope is statistically significantly different from zero at better than 99% confidence level. For 1993 – 2001, the slope is $+1.2$ ppb/year but with a very wide 95% confidence interval of $(-3.2$ ppb/year to 5.6 ppb/year) owing to the extreme variability during this period. For the entire 1980 – 2001 period, the least squares fit gives a slope that is near zero (0.28 ppb/year).

Trends in the annual 95th percentiles of the daily maximum 8-hour ozone concentrations are shown in **Figure 3-4**. The trend in the 95th percentiles is very similar to that in the annual 4th highest value described above. This suggests that the trends in annual 4th highest value described above are indicative of trends in high 8-hour averages in general.

¹ Under the rounding conventions specified in the standard, the 1-hour standard is exceeded by a concentration of 125 ppb or greater while the 8-hour standard is exceeded by a concentration of 85 ppb or greater.

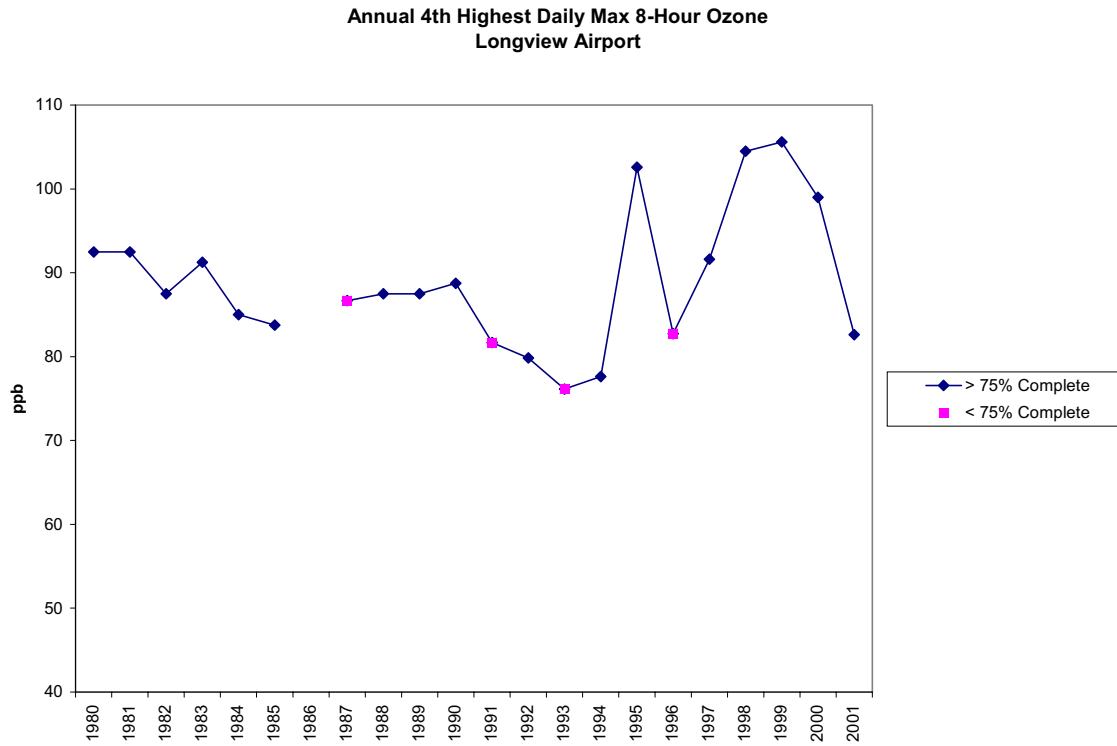


Figure 3-3. Trend in annual fourth highest daily maximum 8-hour average ozone at Longview (CAMS-19).

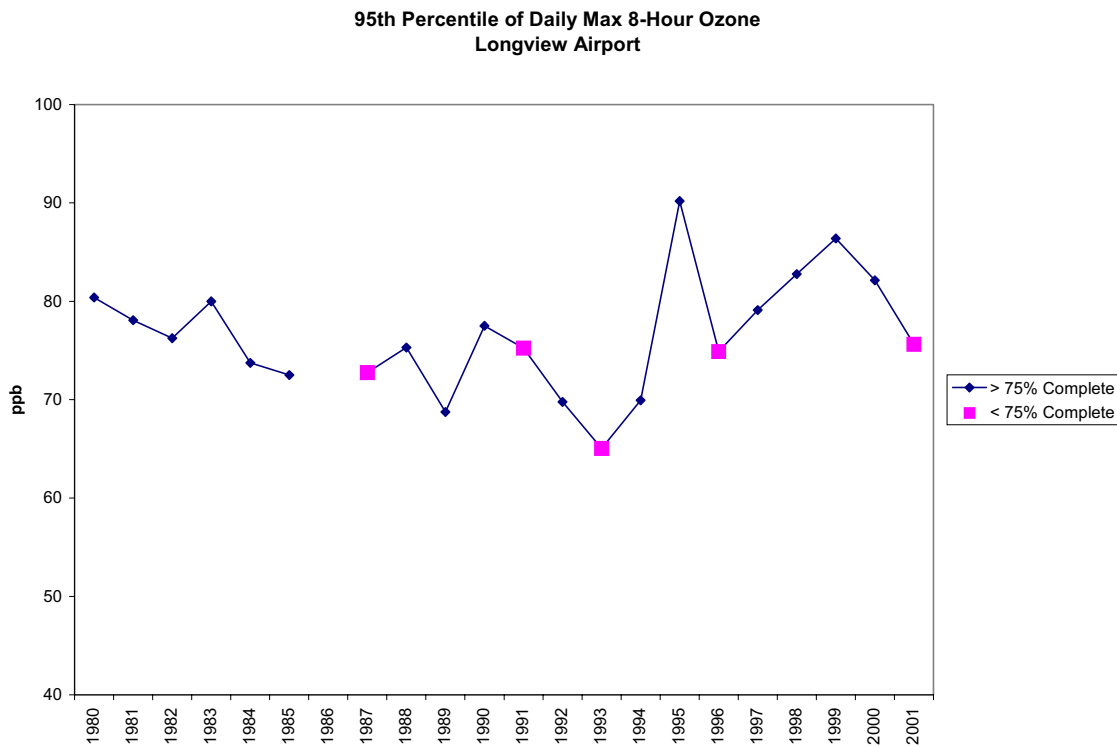


Figure 3-4. Trend in annual 95th percentile of daily maximum 8-hour average ozone at Longview (CAMS-19).

1-Hour Design Value Trends

Annual second highest daily maximum 1-hour ozone concentrations at Longview for the period 1980 – 2001 are shown in **Figure 3-5**. Although the annual 1-hour design values exhibit more year-to-year variability than the 8-hour values, the overall trend is roughly similar, with an overall decrease from 1980 to 1993 or 1994 followed by a period of generally higher values. Trends in the annual 95th percentile of the daily maximum 1-hour averages are shown in **Figure 3-6**. Again, the trend is similar, with less variability than in the annual 1-hour design values but more than in the annual 8-hour design values described above.

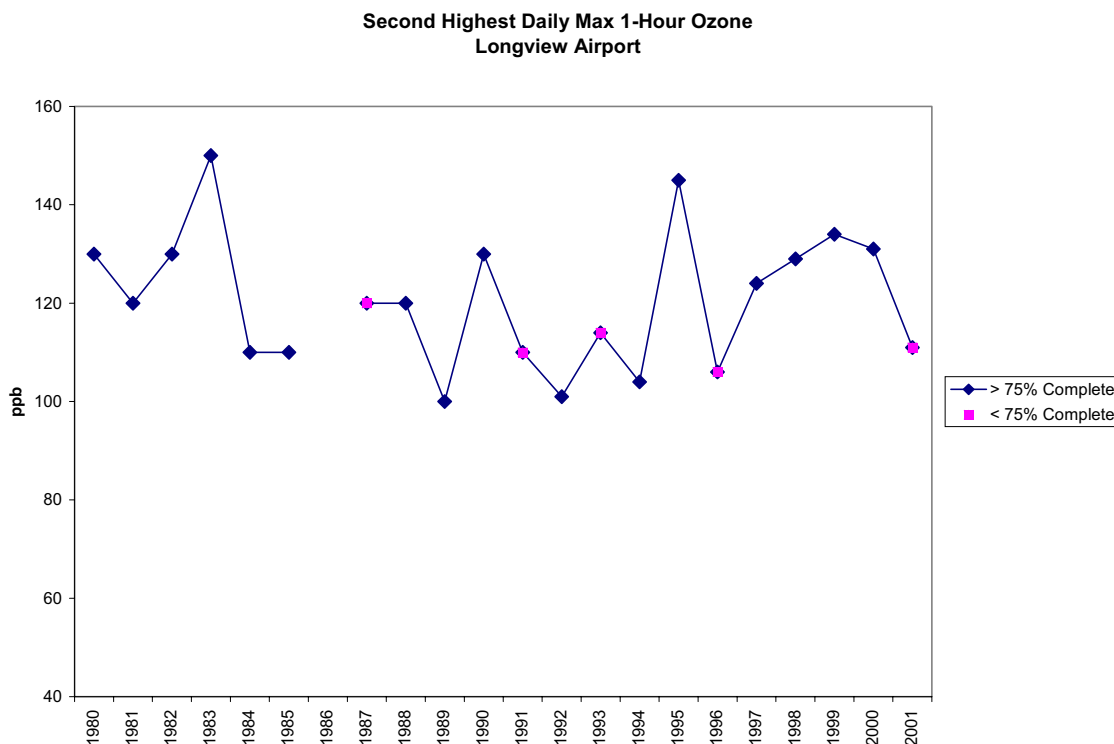


Figure 3-5. Trend in annual second highest daily maximum 1-hour ozone at Longview (CAMS-19).

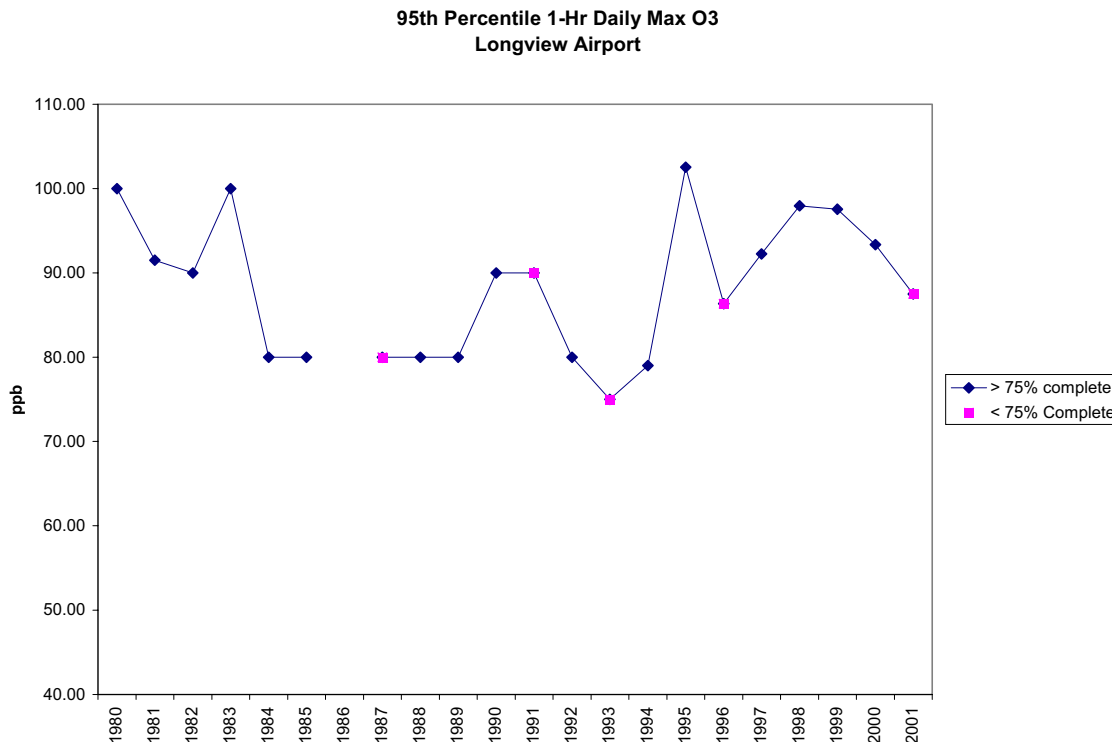


Figure 3-6. Trend in annual 95th percentile of daily maximum 1-hour ozone at Longview (CAMS-19).

Trends in Number of 1-Hour and 8-Hour Exceedance Days

Trends in the number of annual exceedances of the 1-hour standard (i.e., daily maximum 1-hour values greater than 124 ppb) and the number of annual exceedances of the 8-hour standard (i.e., daily maximum 8-hour values greater than 84 ppb) are shown in **Figures 3-7 and 3-8**, respectively. Exceedance counts have been adjusted to account for missing data as per EPA procedures (40 CFR 50, Appendix H). Note that 8-hour exceedances are more common than 1-hour exceedances at Longview, which conforms to the norm. The trend in 8-hour exceedances is very similar to that in the 8-hour design value discussed above: exceedances declined fairly steadily between 1980 and 1994 but then rebounded and followed a much more irregular pattern between 1995 and 2001. More exceedances occurred in 1995 than in any other year. As is the case with design values, the trend in 1-hour exceedances is less obvious. In the case of the exceedances, this is mostly due to the fact that there were very few 1-hour exceedances in any individual year; note that no exceedances were recorded in 1987 – 1989, 1991, 1996, and 2001. Taking these factors into account, the trend in 1-hour exceedances is broadly similar to the trend in 8-hour exceedances, with declining values down to generally none or one per year through 1994 followed by several years with multiple exceedances.

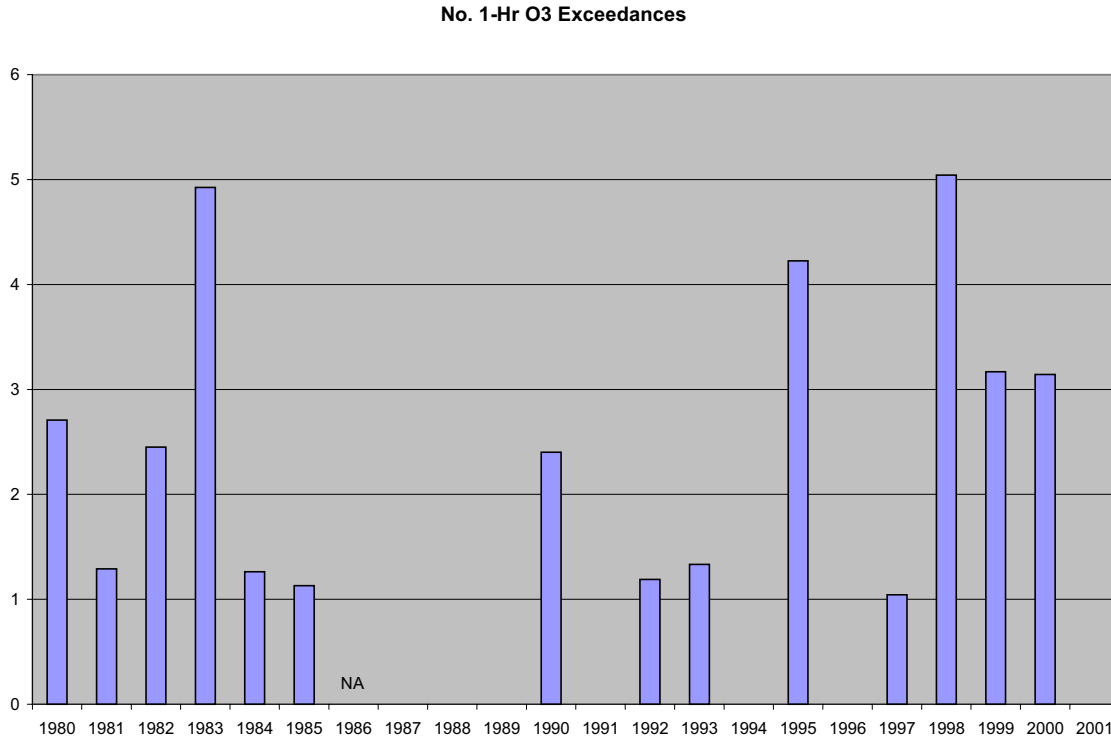


Figure 3-7. Trend in annual number of days exceeding one hour ozone standard at Longview (CAMS-19); values adjusted for missing data.

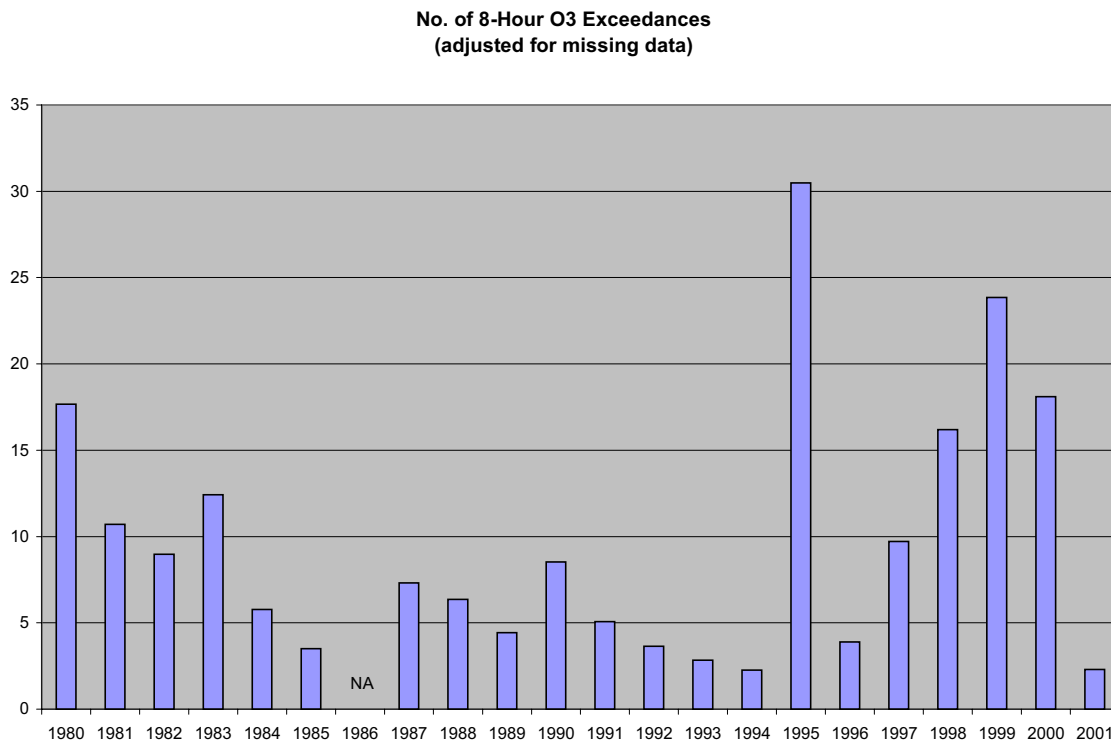


Figure 3-8. Trend in annual number of days exceeding eight hour ozone standard at Longview (CAMS-19); values adjusted for missing data.

Comparison of 1980-1994 and 1994-2001 Trend Periods

As noted above, the year-to-year behavior of annual peak ozone concentrations at the Longview monitoring site appears to be quite different between the early (1980 until approximately 1993) and late (1994 – 2001) periods. This is especially noticeable in the annual fourth highest daily maximum 8-hour ozone concentrations (Figure 3-3) and in the frequency of 8-hour exceedances (Figure 3-8). The apparent change in behavior is not linked to any known specific cause. Review of the emission trends presented at the beginning of this section does not reveal any obvious significant changes in emissions from nearby sources in the mid-1990s which might explain this phenomenon.

In an effort to better understand the apparent change in ozone during the mid-1990s, we compared percentiles of the distribution of daily maximum 8-hour ozone for the early period with corresponding percentiles in ozone from the late period. Only data for May – October were used for this comparison. Results are presented in the form of a quantile-quantile plot in **Figure 3-9**, with the 1:1 line drawn in for reference. Data points falling above the 1:1 line indicate that the quantile corresponding to the data value in the early period (plotted along the x-axis) is lower than the corresponding quantile in the late period data. For example, the 0.5 quantile (i.e., the 50th percentile or median) of the 1980 – 1993 data is 49 ppb while the median for the late period (1994 – 2001) is 54 ppb. An interesting feature of Figure 3-9 is that the difference in quantiles is nearly constant from the lowest values all the way up through about 70 ppb, after which the difference increases. This is shown more clearly in **Figure 3-10** in which we have plotted the differences in quantiles between the early and late periods for quantiles ranging from 0.01 to 0.99. The difference averages about 4.5 ppb up to the median, after which it increases to more than 8 ppb. This is indicative of a greater frequency of high daily max 8-hour concentrations during the late period as compared to the early period. Thus, not only has the whole distribution shifted to the right (i.e., towards higher values), but the right tail of the distribution has become heavier, suggesting a change in the underlying ozone production processes. All else being equal, if emissions were decreasing, we would expect the values of the higher percentiles to decrease but values of the lower percentiles to stay about the same or even increase slightly. Indeed, this is the case when comparing the data from 1980 – 1982 with 1991 – 1993 (not shown). Increasing emissions should have the opposite effect, but the across the board shift to higher values seen in Figure 3-9 is not the expected pattern if emissions had been increasing. It is highly unlikely that this shift is associated with any systematic change in meteorological conditions, given the length of each sub-period (14 years in the early period and 8 years in the late period). This is confirmed in the case of daily maximum temperatures by constructing the appropriate quantile-quantile plot for temperatures (see **Figure 3-11**). As we would expect, there is no systematic difference in the distribution of daily maximum temperatures between the two periods: the data fall close on either side of the 1:1 line. Of course, other meteorological factors can influence ozone levels at Longview, but not only is it very unlikely that any systematic change in other meteorological conditions large enough to have produced the observed change in ozone occurred between the early and late periods, it is even more unlikely that, had such a change occurred, it would not also have shown up in the temperature record.

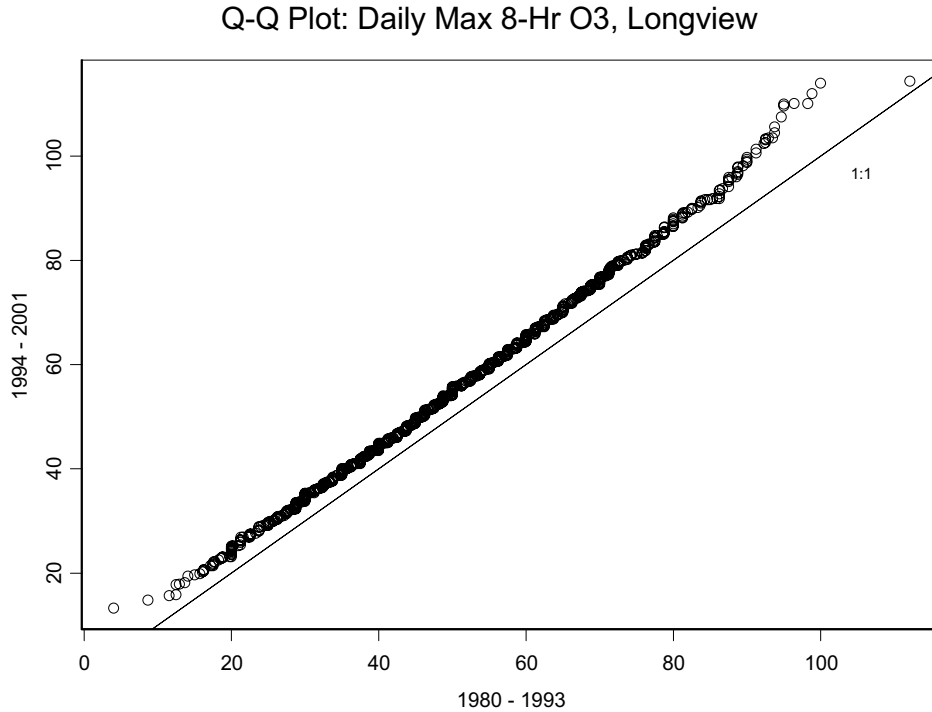


Figure 3-9. Quantile-quantile plot for daily maximum 8-hour average ozone distributions at Longview from 1980 – 1993 data versus 1994 – 2001 data (axis units are ppb).

Differences in Quantiles

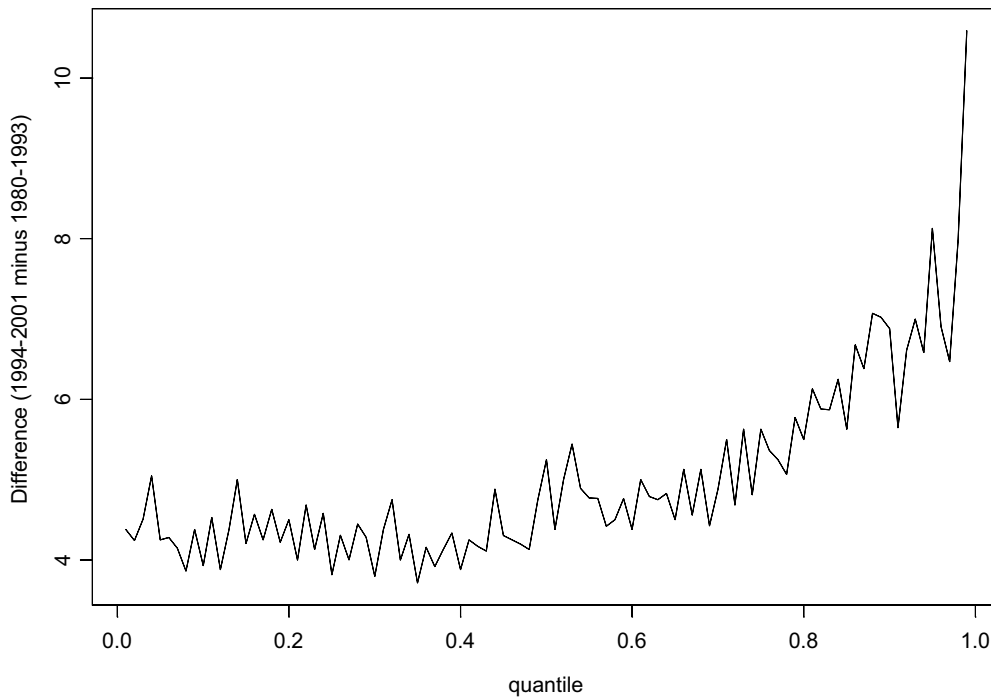


Figure 3-10. Differences (ppb) in quantiles of daily maximum 8-hour average ozone concentration distributions between 1980 – 1993 and 1994 – 2000.

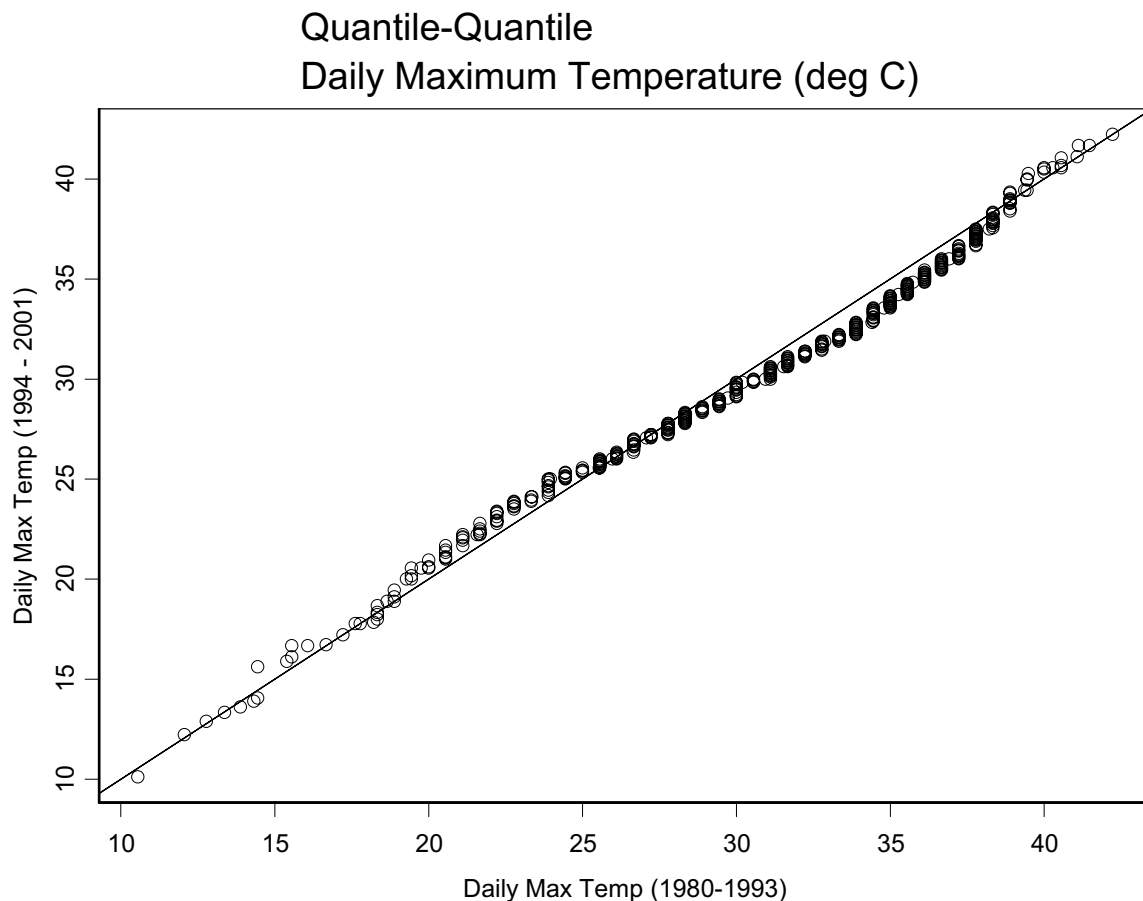


Figure 3-11. Quantile-quantile plot of daily maximum temperature distributions from 1980 – 1993 data versus 1994 – 2001 data (axis units are in degrees Celsius).

Analysis of 1-Hour to 8-Hour Ratios

Given the proximity of the Longview monitor to large NO_x point sources, high ozone levels may be associated with ozone generated along the margins of NO_x plumes originating from one or more of the major combustion point sources in the area (or within a combined NO_x/VOC plume) rather than a broader scale ozone cloud generated by emissions from more widely spread mobile and area sources or distant sources. On warm, light wind days, with plenty of highly reactive biogenic VOCs available, the formation of significant local scale ozone peaks in association with local plumes appears plausible. In addition, some sources in the vicinity of the monitor (e.g., Texas Eastman) emit both NO_x and VOCs. A signature characteristic of this ozone formation mechanism would be observation of short-lived ozone peaks at the fixed monitoring site which would express themselves as relatively high daily ozone “peak to mean” ratios.

A readily available measure of the peak-to-mean ratio can be obtained by dividing the daily maximum 1-hour ozone concentration by the daily maximum 8-hour concentration. A summary of these 1-hour peak to 8-hour peak ratios is presented in the form of annual box plots in **Figure 3-12**. Ratios were computed only for days with daily maximum 8-hour ozone

greater than 70 ppb so as to focus attention on days with significant ozone levels. Ratios vary widely from day to day with median values averaging about 1.14; there is no obvious trend over time. However, when comparing ratios for Longview with ratios for other sites, the Longview monitor stands out in that it has a greater frequency of high ratios. This is illustrated by the quantile-quantile plot shown in **Figure 3-13**. Deviation of the data to the right of the 1:1 line clearly shows the greater frequency of high ratios at Longview compared to other sites. This result confirms an extra degree of “peakiness” of ozone at Longview which is consistent with impact from one or more relatively local point sources.

Peak 1-Hr to Peak 8-Hr Ratios

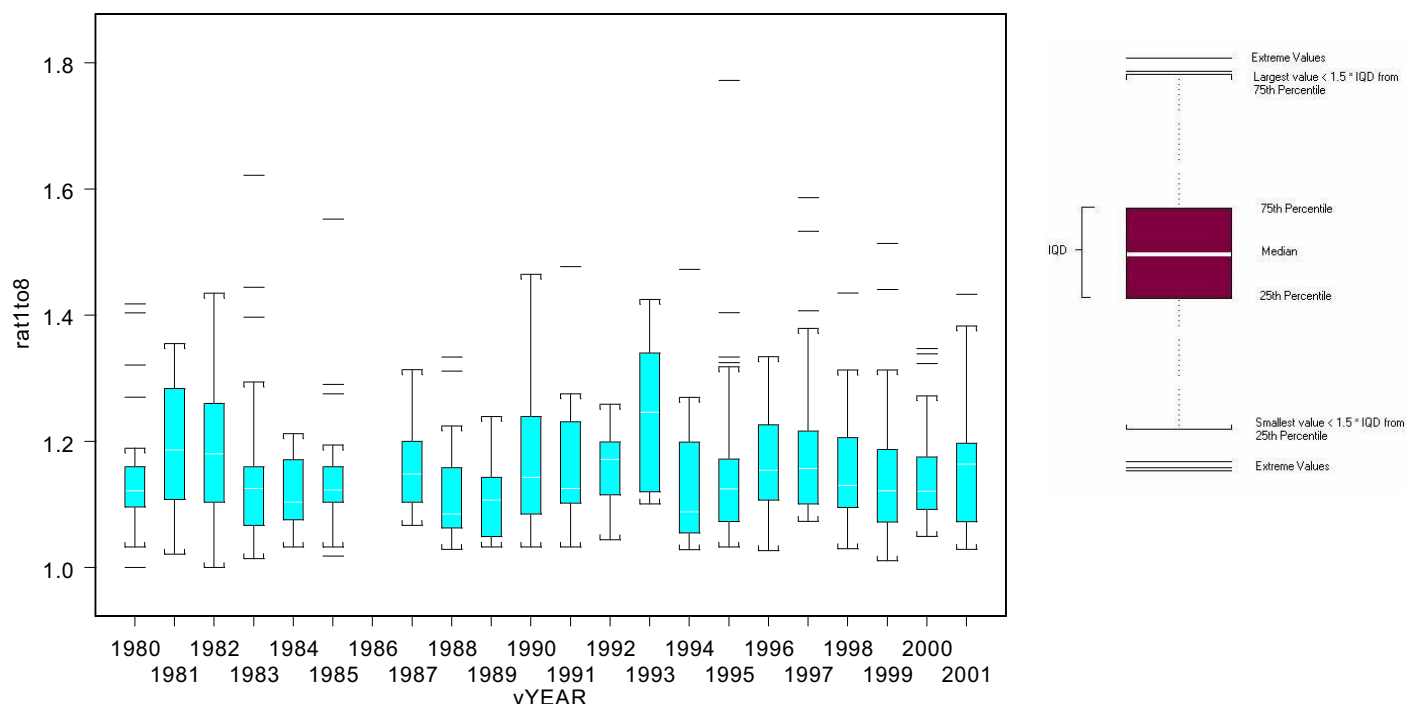


Figure 3-12. Box plots summarizing annual distributions of ratios of daily maximum 1-hour to daily maximum 8-hour average ozone (rat1to8) at Longview (CAMS-19) on days with 8-hour max greater than 70 ppb.

**Quantile-Quantile Plot of Ratios of Daily Max 1-Hr to Daily Max 8-Hr:
1995 - 2001**

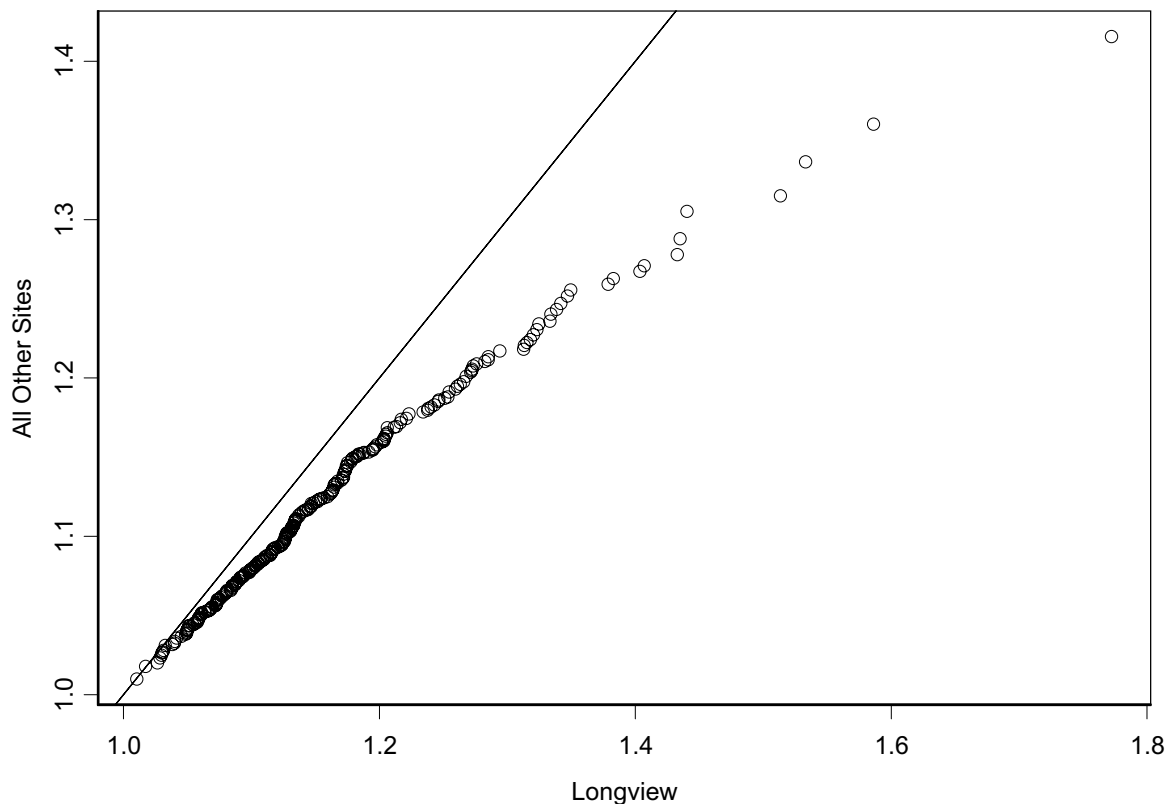


Figure 3-13. Quantile-quantile plot of ratios of daily maximum 1-hour to daily maximum 8-hour ozone at Longview versus all other sites in Northeast Texas and Shreveport, LA.

Comparison with Nearby Ozone Monitoring Data

Trends in annual fourth highest daily maximum 8-hour ozone concentrations at all available monitoring sites with at least 75% complete data in Northeast Texas and neighboring Shreveport, LA are compared in **Figure 3-14**; trends in the annual second highest daily maximum 1-hour concentrations are shown in **Figure 3-15**. Longview is the only site in East Texas with data prior to 1995 so only the 1995 – 2000 period is shown. Longview experienced the maximum value out of all reporting stations in the region in every year except 1996 for both 1-hour and 8-hour ozone annual design values. However, concentrations at the Bossier Parish monitor in central Shreveport have increased steadily over this period and appear to be approaching values seen at Longview. Overall, the trend in ozone values at Longview is consistent with other sites in the region.

Ozone trends for the 1990 – 1999 period at Longview are compared with Dallas and Shreveport in **Figure 3-16**. Annual 8-hour design values for Dallas and Shreveport were obtained from EPA's 2000 Air Quality Trends report (www.epa.gov/airtrends/reports.html) and represent the maximum design value over all sites in the metropolitan area meeting EPA's completeness criteria for trends sites. Note that some ozone monitors in Dallas do not meet these completeness criteria and are therefore not included in EPA's summary. Concentrations

in Dallas track the Longview monitor fairly closely with both locations showing a slight positive trend as illustrated by the linear regression results in **Table 3-1**. The linear trend at Shreveport is closer to zero.

Table 3-1. Linear trends in 8-hour ozone from 1990-1999 from least squares regression.

	Trend	95% Conf. Interval
Longview	+2.38 ppb/year	(0.37, 4.38)
Dallas	+1.55 ppb/year	(-0.75, 3.84)
Shreveport	+0.38 ppb/year	(-0.73, 1.49)

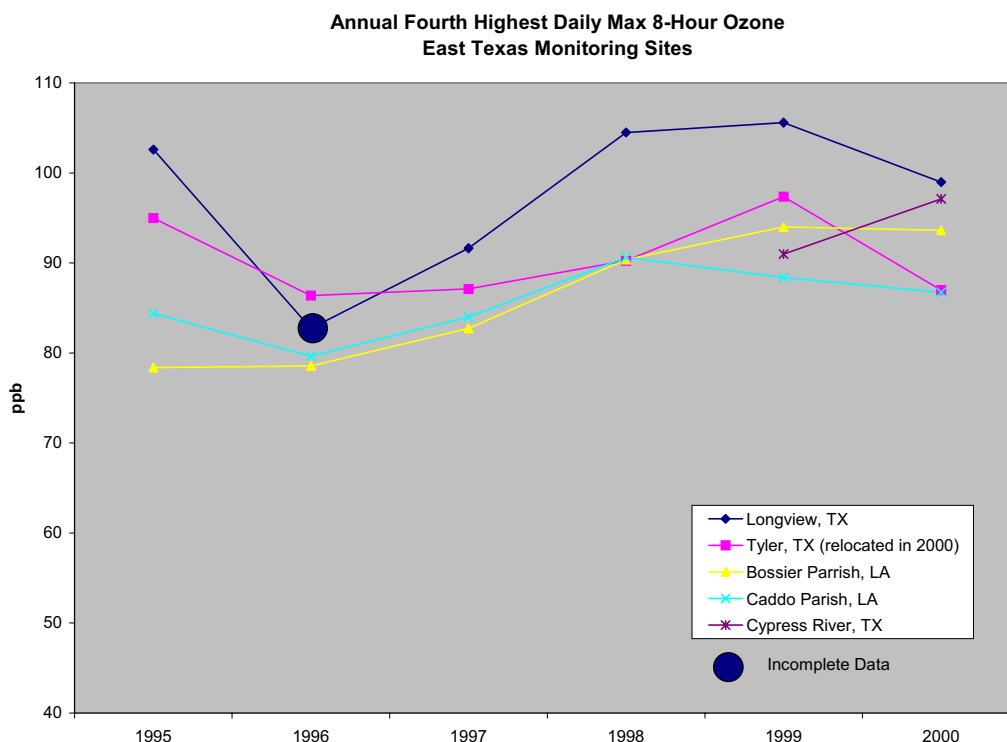


Figure 3-14. Annual fourth highest daily maximum 8-hour average ozone at locations in Northeast Texas and Shreveport, LA.

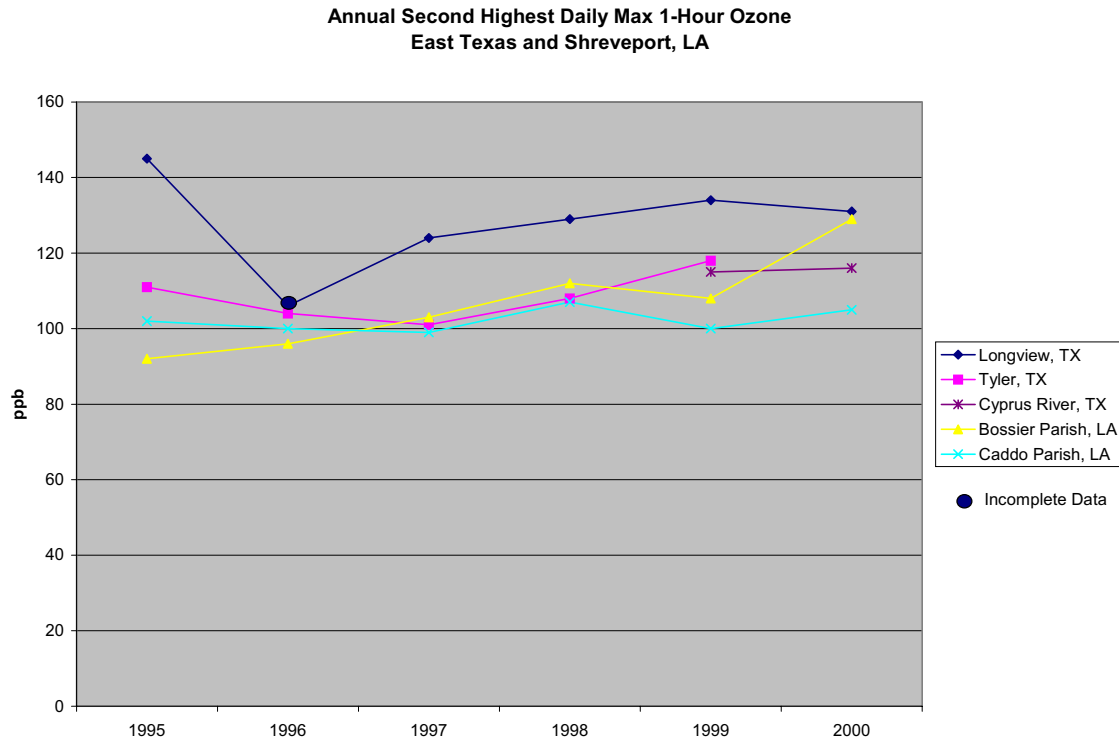


Figure 3-15. Annual second highest daily maximum 1-hour average ozone at monitors in Northeast Texas and Shreveport, LA.

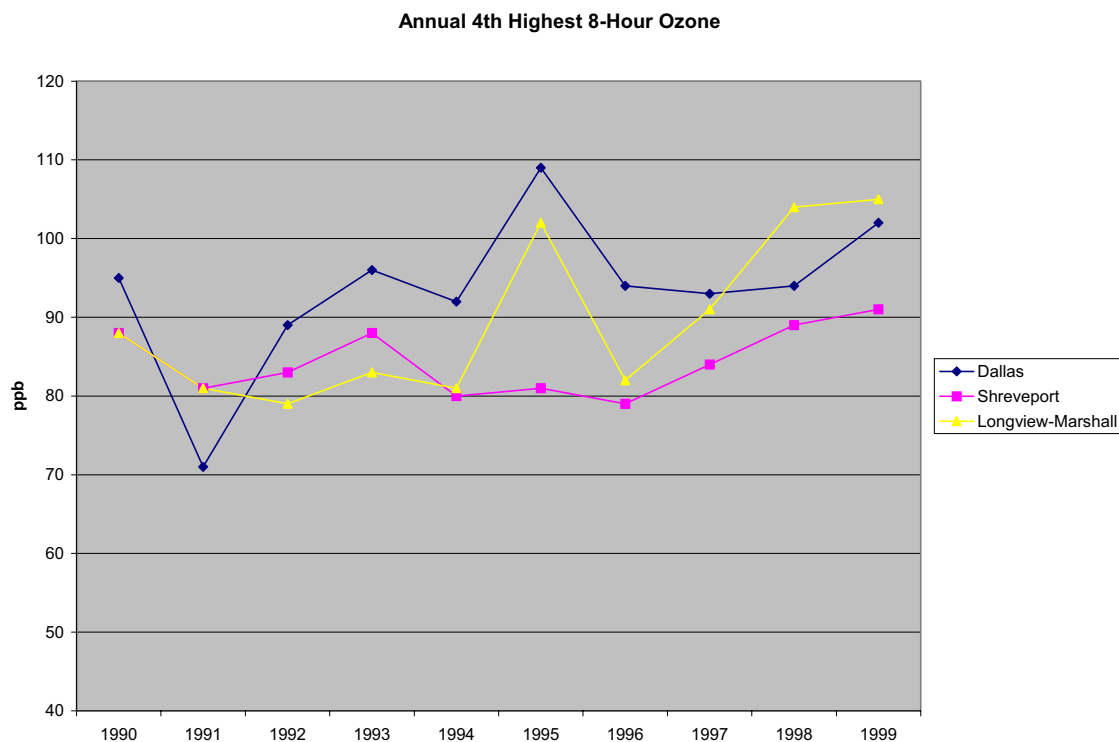


Figure 3-16. Trend in annual fourth highest daily maximum 8-hour average ozone in Northeast Texas, Dallas and Shreveport, LA.

Analysis of Hourly Ozone Data

Patterns in hourly ozone concentrations and correlations with other pollutants were examined to gain further insight into the factors controlling peak ozone levels in the study region. Results of these analyses are presented in the following subsections.

Hourly Ozone and Sulfur Dioxide

A sulfur dioxide monitor has been co-located with the ozone monitor at Longview since 1999. Data from this monitor provide an opportunity to examine the relationship between SO₂ (as a tracer of large fuel combustion sources such as utility and industrial boilers) and ozone during ozone exceedance events. Hourly average ozone and SO₂ data were plotted for all days since 1999 with daily maximum 1-hour ozone exceeding 124 ppb. Results are presented in **Figure 3-17**. On all days, peaks in ozone concentrations occur in conjunction with peaks in SO₂. A particularly good illustration of the close relationship between ozone and SO₂ at this site is provided by the plot for 1 September 2000. There were three daytime peaks in ozone on 1 September, 2000 and all three occurred in conjunction with peaks in the SO₂ concentration. Although the hourly average SO₂ concentrations are relatively low, this is not inconsistent with one or more traverses of the monitoring site by plumes from one or more nearby sources.

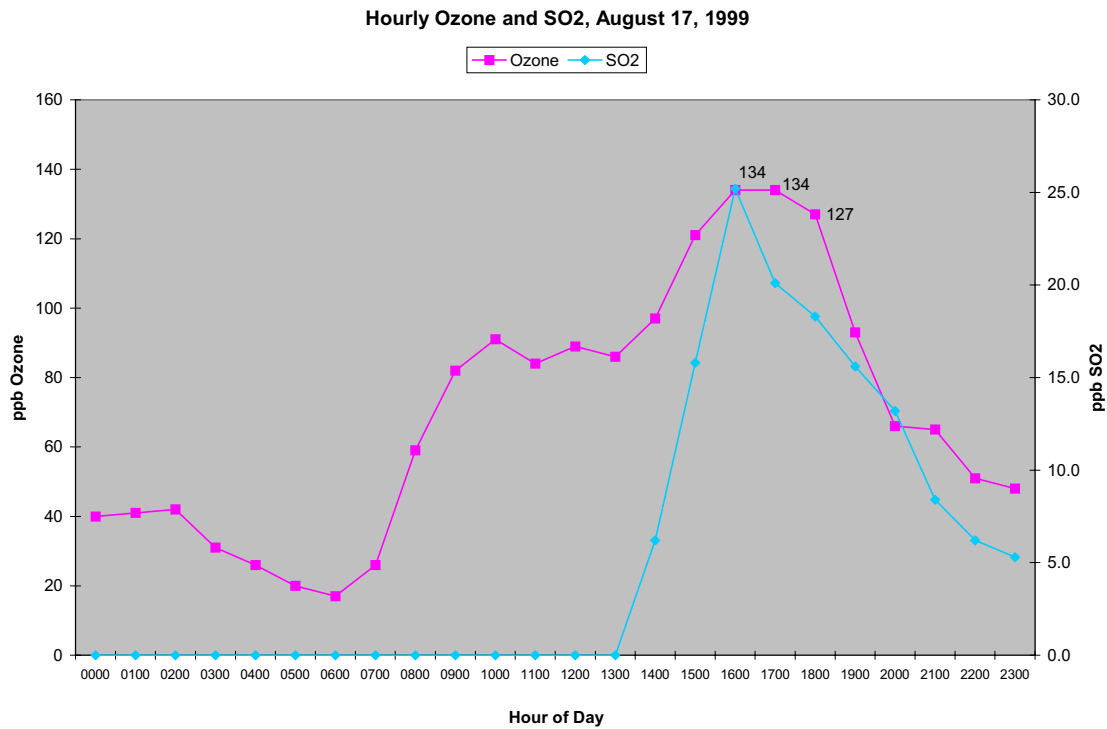
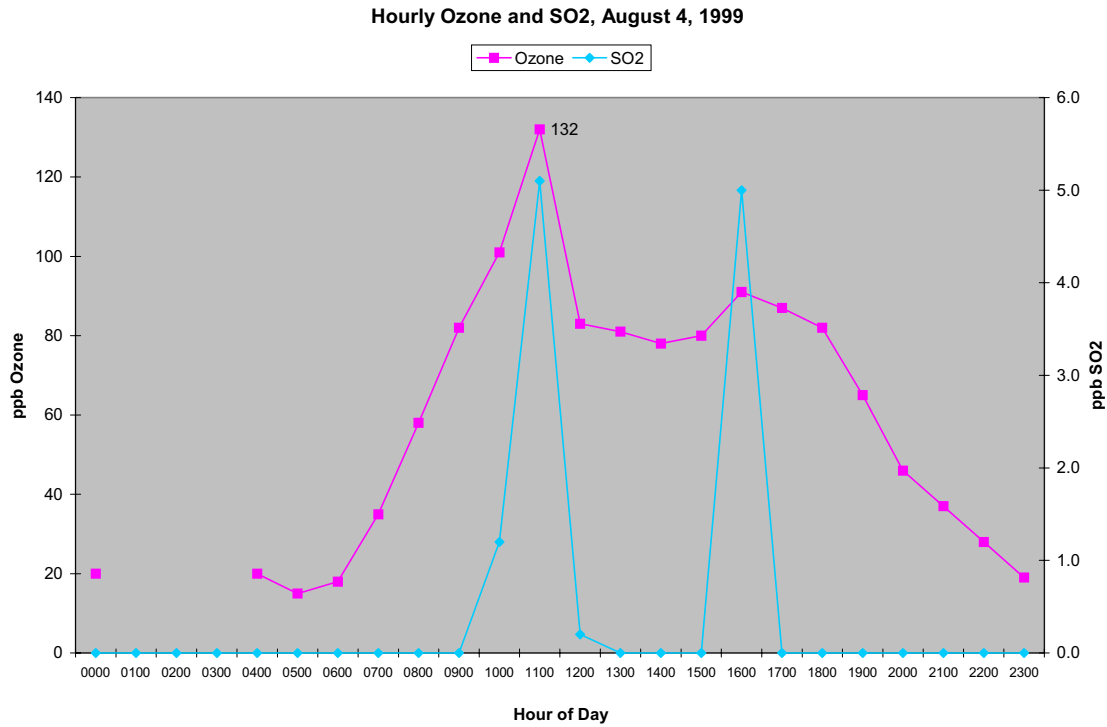
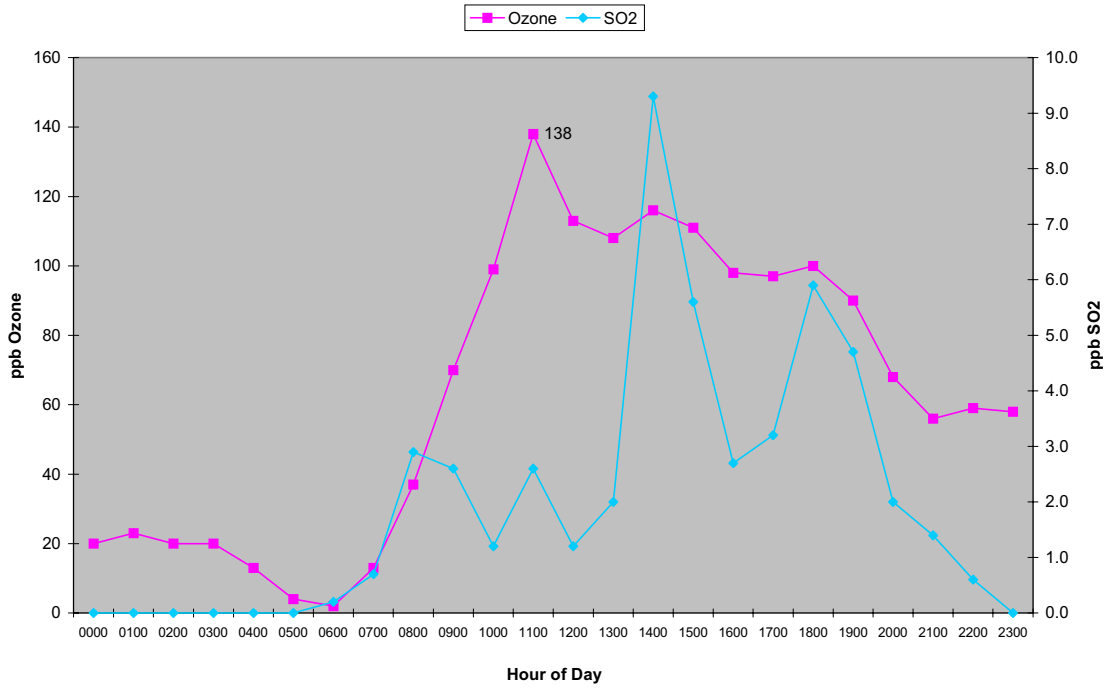


Figure 3-17. Hourly average ozone and sulfur dioxide at Longview (CAMS-19) on days exceeding 124 ppb ozone.

Hourly Ozone and SO₂, September 20, 1999



Hourly Ozone and SO₂, July 15, 2000

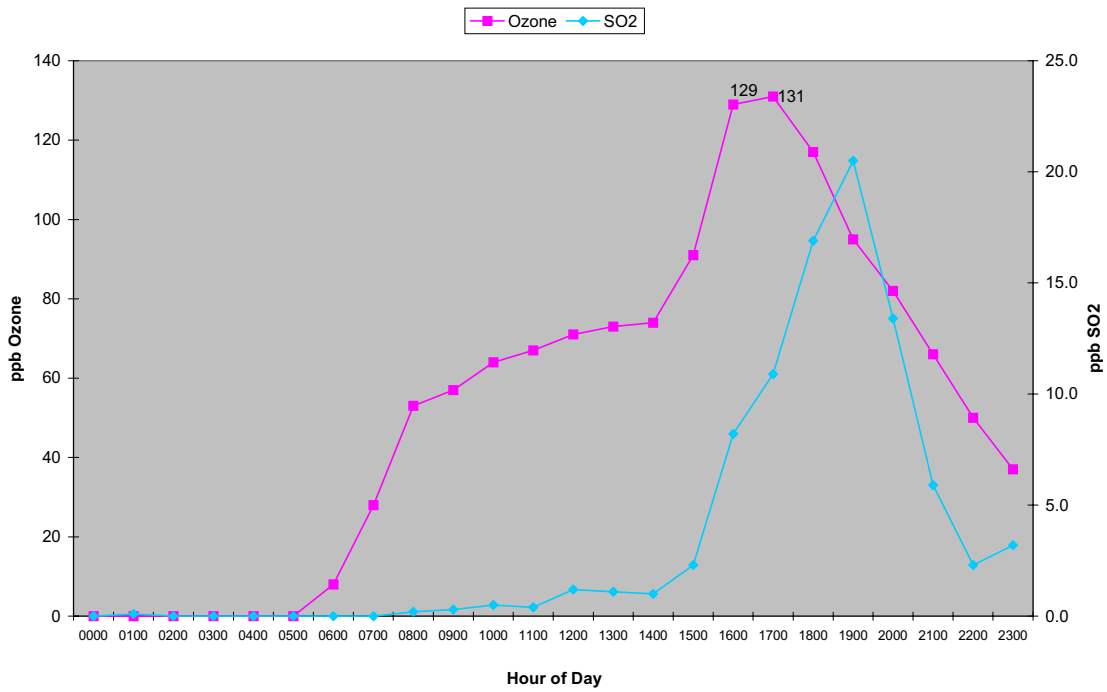


Figure 3-17. continued.

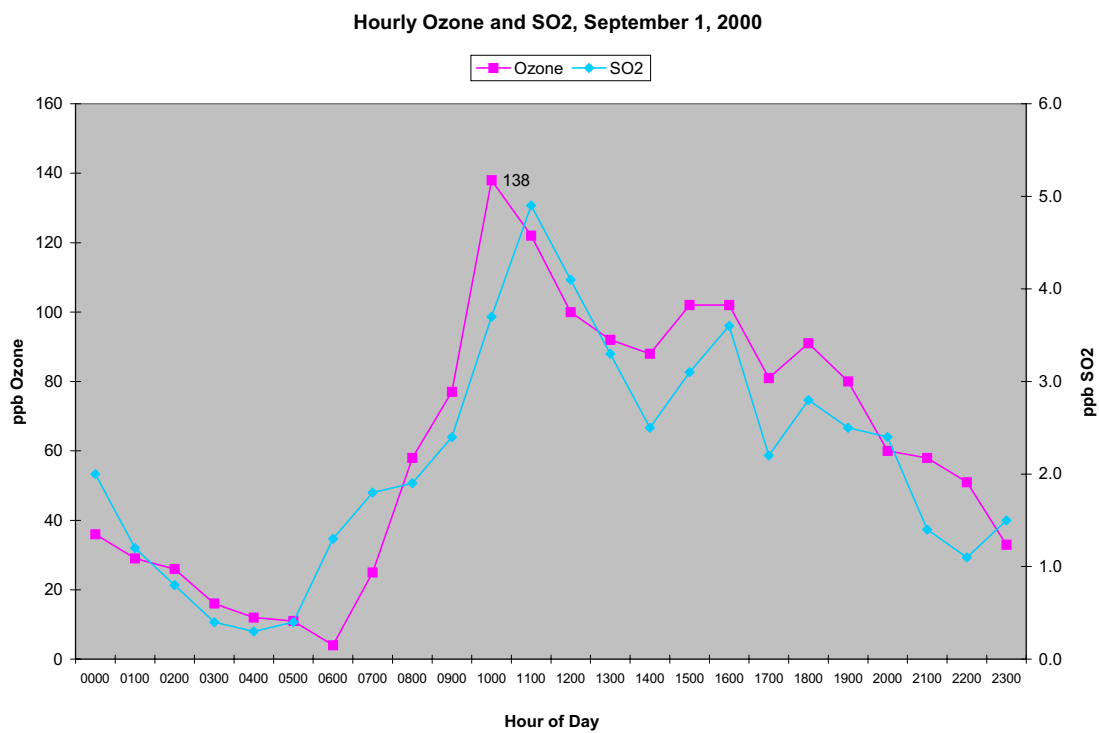
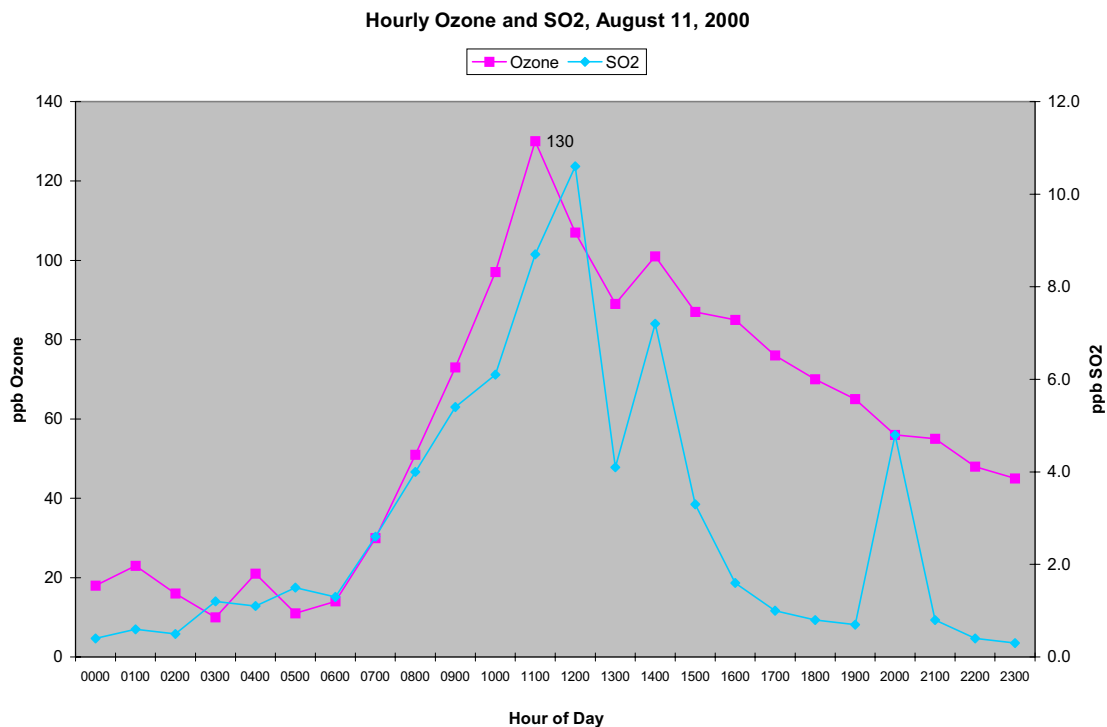


Figure 3-17. Concluded.

Average Diurnal Profiles for Ozone

We attempted to gain additional insight into factors influencing ozone formation and how they might differ over time at Longview by examining average diurnal profiles for ozone on high ozone days. Hourly concentrations were averaged by time of day for 8-hour exceedance days (days with maximum 8-hour averages greater than 84 ppb) between June and October for two sets of years: 1988-1990 and 1998-2000. These years were chosen to examine possible differences in diurnal patterns between the years just before the 1993-1994 minimum in the 8-hour design value trend (see Figure 3-3) and the most recent period with elevated ozone levels. Results are presented in **Figure 3-18**. Error bars in this figure represent 95% confidence intervals for the mean computed from the usual Student's t statistic. These results show an ozone peak in the late afternoon during the 1998-2000 period that is not present in the earlier period. Ozone production is very similar in both periods from the early morning minimum through the 14:00 CST observation. Beginning in 15:00 CST, however, there appears to be formation of extra ozone, peaking at 16:00 CST at which time the average difference is just over 13 ppb. This extra increment of ozone is then maintained until 23:00 CST. As indicated by the non-overlapping error bars, hourly means are statistically significantly different at the 95% confidence level between the two sets of years during this time of the day. It is not clear what might be causing this late afternoon peak - further analyses is need. One plausible explanation, however, is the injection of additional fresh NO into the atmosphere during the middle of the day, causing additional production of ozone as the afternoon progresses.

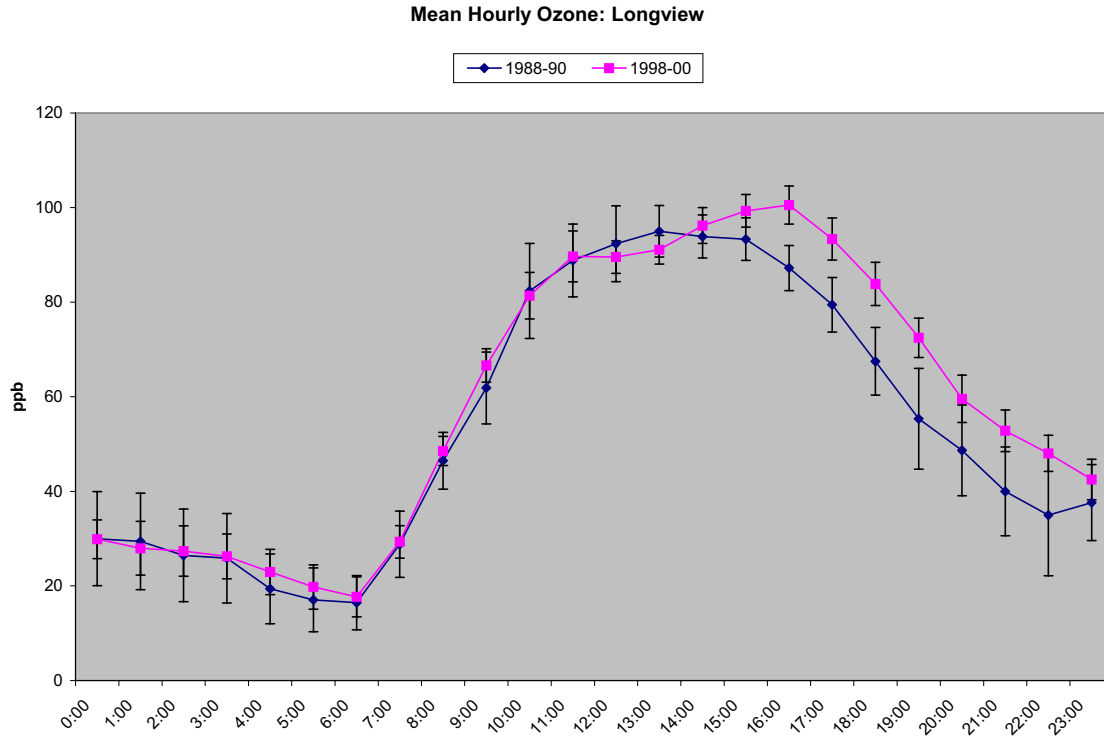


Figure 3-18. Mean diurnal profile of hourly average ozone at Longview (CAMS-19) for 1988 – 1990 versus 1998 – 2000 (error bars indicate 95% confidence intervals).

4. SUMMARY, CONCLUSIONS AND RECOMMENDATIONS

In the preceding section we presented results of an exploratory analysis of trends in ozone and ozone precursor emissions in the Tyler-Marshall-Longview Near Nonattainment Area in Northeast Texas. Our retrospective analysis of air quality and meteorological conditions in this area was complicated by a lack of long-term monitoring data. Prior to 1995, ozone measurements were available only at the Longview monitor (CAMS-19). Historical ozone precursor data are also extremely limited in this region and long-term precursor trend data are not available. This situation has improved in recent years with the establishment of additional permanent monitoring sites at the Tyler airport (CAMS-86) and the Cypress River airport (CAMS-50)/Karnack (CAMS-85). In addition, a special ozone and ozone precursor monitoring program was undertaken at the Longview site during the 2001 ozone season, including collection of 100 two-hour canister samples on 15 separate days for speciated hydrocarbon analysis (AQSI, 2002). This program also included collection of ozone and NO_x monitoring data at one other research site (Big Woods, CAMS 605). In 2002, the research site was moved from Big Woods to Waskom and aircraft sampling was conducted on selected days. Analyses of these data were beyond the scope of our study but should provide further insight into ozone formation mechanisms in the region.

While a detailed emissions inventory covering the NNA for 1999 has been prepared for SIP development purposes, only a limited amount of information is available for estimating emission trends prior to 1999. We assembled what data are available for key source categories: on-road mobile, non-road mobile, and area and major point sources. Data on trends in area and non-road emissions prior to 1990 are currently unavailable.

Our exploratory analysis of ambient air quality, meteorological, and emissions data from the Tyler-Marshall-Longview NNA has identified a number of key features:

- Peak ozone concentrations at Longview declined steadily from 1980 until the mid-1990s. During 1980-1993, the annual fourth highest daily maximum 8-hour average decreased at rate of just under 1 ppb/year (least-squares fit linear trend). From the mid-1990s until 2001, however, ozone design values have fluctuated widely, reaching levels at or above the highest observed throughout the 1980 – 1994 period. This change in behavior during the mid-1990s is most noticeable in the 8-hour design value and exceedance count trends but can also be seen in the 1-hour trends.
- The distribution of daily maximum 8-hour ozone concentrations for 1994-2001 differs from the distribution for 1980-1993 not only in that there is a greater frequency of high concentrations in the later period, but also in that even days without elevated ozone (i.e., those near the median or even at lower percentiles) had slightly higher concentrations in the later period. Reasons for this wholesale shift in the distribution of daily maximum ozone are not clear at this time.
- Emissions of NO_x and VOC in the region declined fairly steadily between 1990 and 2001 with reductions in anthropogenic VOCs exceeding the NO_x reductions on a percentage basis. On-road mobile source NO_x and VOC emissions also decreased between 1980 and 1990 but data on emission trends from most other sources during this period are currently

unavailable. There are no known significant deviations in this trend during the mid 1990s that might explain the change in ozone design values noted above.

- Analysis of ratios of daily maximum 1-hour to daily maximum 8-hour average concentrations on days with elevated ozone (8-hour averages exceeding 70 ppm) reveals that higher ratios are more frequent at Longview than at other monitoring sites in the study region. This result is consistent with the hypothesis that ozone at Longview is subject to the influence of one or more local point sources.
- Comparison of average diurnal patterns in ozone concentrations on 8-hour exceedance days during 1988-1990 and 1998-2000 indicates a period of enhanced ozone production in the late afternoon during the latter period which was not present during the former. This difference is particularly striking because the mean profiles are nearly identical during the rest of the day. Although the reason for this difference is not presently clear, this result is consistent with the availability of more fresh NO at mid-day during the 1998-2000 period. Analysis of additional years of ozone data, along with available NO and NO₂ data is recommended.
- Examination of hourly ozone and sulfur dioxide concentrations at Longview on 1-hour exceedance days shows that ozone peaks are frequently associated with elevated SO₂ levels, providing further evidence of ozone impacts associated with plumes from nearby combustion point sources. More analysis of the O₃ and SO₂ data will be needed to confirm this finding. In particular, given the short time scales associated with plume impacts, we recommend that an analysis of the 5-minute monitoring data be carried out.

While the results summarized above are not yet sufficient to allow us to reach any final conclusions about what factors are driving ozone trends in the Tyler-Marshall-Longview NNA, they provide much evidence in support of the hypothesis that peak ozone events at Longview in particular are frequently associated with impacts from one or more nearby major point sources. Analyses of data from this region prepared by the TNRCC (Sullivan, 2001) have reached similar conclusions. We understand that the TNRCC group is continuing to investigate this issue and will be preparing additional reports the results of which are not currently available.

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Appendix A
Biogenic Emissions

ENVIRON

MEMORANDUM

To: Mark Sweeney, ETCOG

From: Greg Yarwood, Gerry Mansell and Steve Lau

Date: Revised, August 15, 2002

Subject: Biogenic Emissions for Inclusion in the 1999 East Texas Emission Inventory

This memorandum updates a previous June 19, 2001 memorandum of the same title to correct errors in Table 1. There is little change to total biogenic VOC emissions (920.52 compared to 907.25) and no change to the final conclusions. This change has no impact at all on any ozone modeling results for East Texas.

Summary

ENVIRON and Pollution Solutions are preparing a 1999 emission inventory for the 5 county area of East Texas that is part of North East Texas Air Care (NETAC). This emission inventory will be submitted to the Texas Natural Resource Conservation Commission (TNRCC) and also will be used in ozone modeling for East Texas. This memorandum presents the biogenic emissions estimates for the 5 county area (Table 1), describes the methodology and assumptions for the emissions calculation, and compares the present inventory to the previous biogenic emission inventory (Yarwood et al., 1999a).

Table 1. Biogenic emissions (tons/day) for a typical summer day.

County	VOC	NO _x
Gregg	57.09	0.17
Harrison	262.85	0.42
Rusk	239.17	0.40
Smith	224.71	0.59
Upshur	136.70	0.41
Total	920.52	2.01

The new biogenic emissions are based on:

- Version 2.2 of the GloBEIS biogenic emissions model (Yarwood et al., 1999b and 1999c; Guenther et al., 1999a and 1999b) with the “BEIS99” emission factor algorithm.
- The latest landuse/landcover (LULC) data for biogenic emissions developed by the TNRCC (Yarwood et al., 1999b; Wiedinmyer et al., 1999).

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- Weather conditions for a typical summer day in East Texas in 1999.

The new inventory supersedes the previous biogenic emission inventory prepared for East Texas in 1998 (Yarwood et al. (1999c). The new emission totals are consistent with the assumptions in the final round ozone control strategy modeling for 1-hour ozone performed in 1999, so it is not expected that these updates to the biogenic inventory would change the control strategies for East Texas.

The emission totals presented in Table 1 are for a typical summer day condition. Biogenic emission inventories for ozone modeling will be re-calculated for the temperatures that actually occurred on the days being modeled (i.e., the biogenic emissions will be day specific) using the same model and LULC data.

GloBEIS2 Model and Input Data

Emissions were calculated using Version 2.2 of the GloBEIS biogenic emissions model (Yarwood et al., 1999b and 1999c;). GloBEIS2 includes options for calculating emissions using either the BEIS2 methodology or the updated BEIS99 methodology (Guenther et al., 1999a and 1999b). The BEIS99 option was used here because it represents improved science over BEIS2. GloBEIS also includes an option to adjust the BEIS99 emissions for seasonal variations in biomass density. The seasonal adjustment was not used because the TNRCC would like to further review the methodology before using it in emission inventories.

GloBEIS requires external data to describe the landuse/landcover, temperatures and solar radiation. The landuse/landcover (LULC) data were from the most recent study sponsored by the TNRCC (Yarwood et al., 1999b; Wiedinmyer et al., 1999).

Temperature and Cloud Cover Data

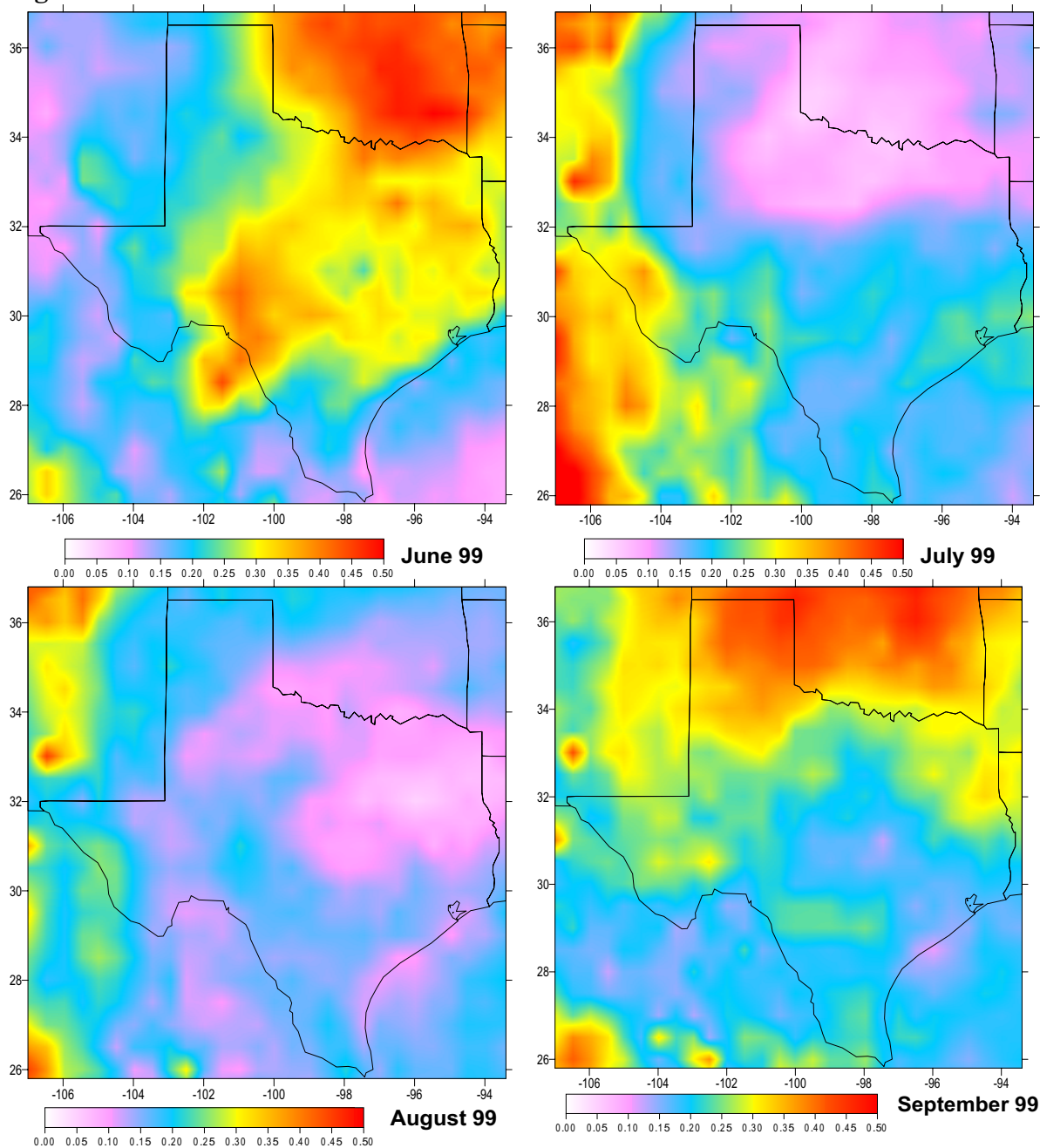
Representative temperature and cloud cover data for an ozone season day were developed as part of a TNRCC project to improve the 1999 emission inventory for all counties in Texas. The objective was to characterize typical conditions on a high ozone day in 1999, not to characterize any one particular day.

Surface temperatures were characterized by averaging over two periods (August 13-22, and September 13-20, 1999) when high ozone levels were observed in many parts of Eastern Texas. These periods are also the basis of photochemical modeling currently being developed for several ozone “near nonattainment areas” so the county level biogenic emissions developed here are generally representative of biogenic emissions being used in the 1999 ozone modeling. Hourly surface temperature observations archived by the National Climatic Data Center (NCDC) were analyzed to develop an average temperature profile for each county, as shown in Table 2. These temperature data are further described in the report on the preparation of the biogenic emission inventories for ozone modeling (Yarwood et al., 2001).

Table 2. Average hourly temperatures (K) for the 5 counties during summer 1999 (in Kelvin).

	0	1	2	3	4	5	6	7
Gregg	297.1	296.4	295.8	295.1	294.4	294.0	293.7	295.1
Harrison	296.9	296.1	295.5	294.8	294.2	293.7	293.4	294.9
Rusk	297.0	296.2	295.7	294.9	294.2	293.8	293.6	295.1
Smith	297.7	296.9	296.3	295.6	294.9	294.5	294.2	295.4
Upshur	297.2	296.4	295.8	295.2	294.5	294.0	293.7	295.1
	8	9	10	11	12	13	14	15
Gregg	297.7	300.1	302.0	303.7	305.0	306.0	306.9	307.1
Harrison	297.5	300.0	301.9	303.6	304.8	305.8	306.7	306.9
Rusk	297.8	300.4	302.3	304.1	305.4	306.3	307.2	307.4
Smith	298.0	300.3	302.2	304.0	305.3	306.3	307.2	307.4
Upshur	297.6	299.9	301.9	303.6	304.9	305.9	306.8	307.0
	16	17	18	19	20	21	22	23
Gregg	307.1	306.5	305.4	302.7	300.4	299.4	298.6	298.0
Harrison	306.8	306.3	305.1	302.4	300.2	299.2	298.3	297.8
Rusk	307.4	306.6	305.4	302.5	300.1	299.1	298.3	297.7
Smith	307.5	306.8	305.7	303.1	300.9	299.9	299.0	298.4
Upshur	307.0	306.5	305.3	302.7	300.6	299.6	298.7	298.1

Cloud cover was characterized from Geostationary Operational Environmental Satellite (GOES) data analyzed and archived by the University of Maryland (UMD). UMD provided average cloud cover fraction for the months of June, July, August and September 1999 on a half-degree grid covering the continental US. These data are shown for Texas in Figure 1. The cloud cover fraction over Texas in 1999 varied from month to month according to the passage of stormy and clear weather systems. Overall, a cloud cover fraction of two tenths appears representative for summer conditions in East Texas, and so this was used as the basis for calculating the biogenic emissions. Day specific UMD satellite data are used in the preparation of day specific biogenic emission inventories for ozone modeling (Yarwood et al., 2001).

Figure 1. Cloud Fraction Plot for Summer 99.

GloBEIS2 Emissions

Comparison to Previous Inventory

The differences in methodology used for the previous biogenic emissions inventory (Yarwood et al., 1999a) are summarized below and the previous emission totals for the five county area are shown in Table 3.

- Older version of the TNRCC LULC data. The main changes (Yarwood et al., 1999b) were an updated methodology for crop lands.

- Older version of the GloBEIS model. The main update for GloBEIS2 was the inclusion of updated isoprene emission factor algorithms (called BEIS99) in place of the older BEIS2 algorithms.
- Different temperatures. The previous inventory assumed slightly warmer temperatures, on average.
- Previous inventory was derived from a gridded inventory (as used for photochemical modeling). Consequently, the previous emission totals were only available for the total 5 county area and were calculated by adding up the emissions for all grid squares covering the 5 county region.

Table 3. Total biogenic emissions (tons/day) for the five county region from the previous study.

	Total
VOC	1350.2
NO _x	2.00

Comparing the emissions totals in Tables 1 and 3 shows:

- Decrease in the total VOC emissions for the five county area of 32 percent for the new inventory. This is consistent with the changes between the BEIS99 and BEIS2 algorithms.
- No change in NO_x emissions.

The latest biogenic emission totals lead to the following conclusions regarding the role of biogenic sources in ozone formation in East Texas.

- Biogenic VOC emissions are still large relative to anthropogenic VOC emissions for the five county area and will play an important role in local ozone formation.
- Biogenic NO_x emissions are small and will make negligible contribution to local ozone formation.

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