OXIDANT AND PRECURSOR TRENDS IN THE METROPOLITAN LOS ANGELES REGION

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For Presentation at
The International Conference on Photochemical
Oxidant and Its Control

Raleigh, North Carolina

September 1976

ABSTRACT

This paper describes recent historical trends in oxidant and precursors in the Los Angeles region. Control strategies and basin-wide emission trends for nitrogen oxides and reactive hydrocarbons are documented year by year from 1965 to 1974. Trends in the geographic distribution of emissions are illustrated by computing net percentage emission changes over the decade for individual counties. The changes in emissions are compared to changes in ambient precursor concentrations and oxidant concentrations. It is found that many of the changes in monitored air quality can be explained by trends in total emissions and in the spatial distribution of emissions.

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1. INTRODUCTION

The control of photochemical oxidant is a complex, dynamic process involving major technical uncertainties. An effective way to deal with the dynamic and uncertain aspects of the problem is to adopt a "feedback" approach in control strategy planning. Progress (or lack of progress) in improving ambient air quality should be continually reviewed. Control strategies should be periodically reexamined and, if necessary, reformulated. Control plans should be flexible so that changes can be made as new information is developed.

Trend analysis constitutes a fundamental part of the feedback approach to air pollution control. Air quality and emission trends should be major factors in determining whether or not control strategies need reformulation. Trend studies also can help to reduce technical uncertainties. For instance, the analysis of ambient precursor trends can indicate the adequacies, or deficiencies, in emission inventories and emission reduction estimates. Comparing historical oxidant changes to precursor emission changes can provide insight into the relationship between emissions and air quality. This improved knowledge can be used in evaluating future control strategies.

This paper documents trends for oxidant and the precursors (nitrogen oxides and reactive hydrocarbons) in the Los Angeles region from 1965 to 1974. Changes in basinwide precursor emissions and in the geographic distribution of emissions are compared to changes in ambient precursor concentrations and ambient oxidant levels. Most of the results presented here are based on a recent report of the Caltech Environmental Quality Laboratory [1]. The reader is referred to that report for detailed discussions of the methodology and results.

2. PRECURSOR EMISSION TRENDS

This section describes trends in control strategies and emissions over the last decade for the two oxidant precursors, nitrogen oxides and reactive hydrocarbons. The trend estimates are based on much of the latest information available for emission sources in the Los Angeles region. Included are new reactivity factors for hydrocarbon emissions [2,3], recent measurements of auto exhaust emission and deterioration factors [4,5,6,7,8], test data on the unexpectedly large contributions made by evaporative emissions from light-duty vehicles [9,10], the latest stationary source NO_x inventory [11], and recent data on traffic (VMT) growth and fuel usage patterns [12,13,14,15,16]. A detailed description of the methodology for determining the emission trends can be found in Reference [1].

Basin-wide emissions of each pollutant are documented year by year from 1965 to 1974 in order to characterize overall emission trends and the changing role of various source categories. To illustrate the spatial distribution of emission trends, net percentage changes in emissions over the decade are determined for each of the six counties within (or partially within) the Metropolitan Los Angeles AQCR. Figure 1 shows the area covered by the Los Angeles air basin and the geography of the six county sub-areas.

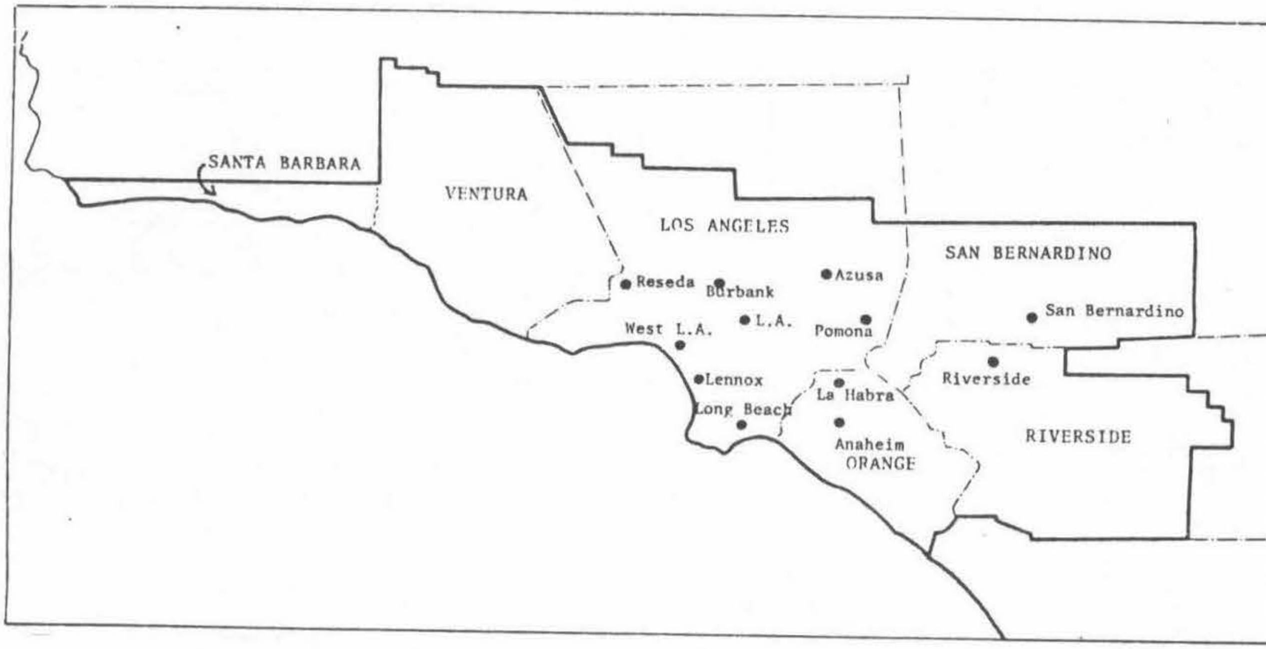

Figure 1. Map of the Metropolitan Los Angeles AQCR

Trends in Total Emissions

The purpose of documenting basin-wide trends in total emissions is to help explain trends in average, basin-wide air quality. Total emission trends are also useful for illustrating changes in emission levels for various source categories. These changes result from emission control actions in competition with the growth of source activity levels. Since different source categories are associated with different rates of growth and degrees of control, the roles of various source categories have undergone continual alteration.

Figure 2 presents our estimates of basin-wide NO_x emission trends over the past decade. The basin-wide total is represented by the top line, while the distances between the other lines illustrate the contributions of various source categories. Figure 2 shows that total NO_x emissions in the Metropolitan Los Angeles AQCR increased by about 36 percent from 1965 to 1974. The major contributor to this increase was the rise in NO_x emissions from light-duty vehicles; total NO_x emissions from light-duty vehicles increased by 75 percent over the decade. This was partially due to a 41 percent increase in basin-wide VMT and partially due to the large increase in NO_x emissions among 1966-1969 model year vehicles. The control techniques used to reduce hydrocarbons and carbon monoxide in 1966-1969 vehicles had the side effects of raising NO_x emissions. New car emission standards for NO_x took effect in 1971, but as yet these standards have not had a great impact on basin-wide NO_x emissions.

Heavy-duty vehicles were also a growing source category for NO_x over the decade. Basin-wide emissions from heavy-duty vehicles increased by 40 per-

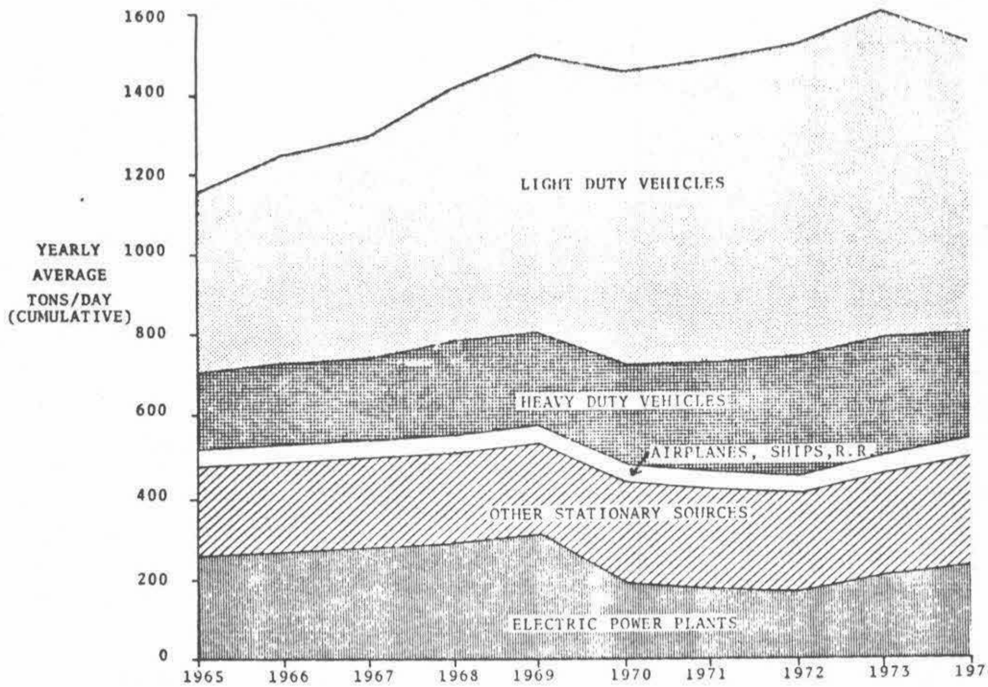

Figure 2. Total NO_x Emission Trends in the Los Angeles Region

cent, basically due to VMT growth. Although facing a high rate of demand growth, the electric power plants have contained and reduced their NO_x emissions effectively by use of natural gas and substitution of low-sulfur fuel for high-sulfur fuel in the sixties and by the Rule 68 retrofits and the industry's own extensive work which caused significant reductions after 1969. Nevertheless, the years 1972-1974 have already witnessed the growth of power plant NO_x emissions due to shortage of natural gas and increased use of fuel oil. Emissions from other stationary sources of NO_x essentially increased with energy use by those sources.

A slight downturn in total NO_x emissions occurred from 1973 to 1974. This downturn is partially due to emission controls on new light-duty vehicles, but it mostly reflects the energy crisis and resulting decrease in traffic from 1973 to 1974. The downward trend in total NO_x emissions should continue at an even greater rate after 1975 because of the stringent new car NO_x emission standards which started in 1975.

Figure 3 summarizes basin-wide trends in reactive hydrocarbon emissions from 1965 to 1974. The top line represents total RHC emissions, and the distances between the lines represent the contributions from various source categories. Here, the definition of reactive hydrocarbon emissions is based on a two-group reactivity scheme proposed by Dimitriades [2] of the Environmental Protection Agency and on reactivity factors calculated in a recent TRW report [3].

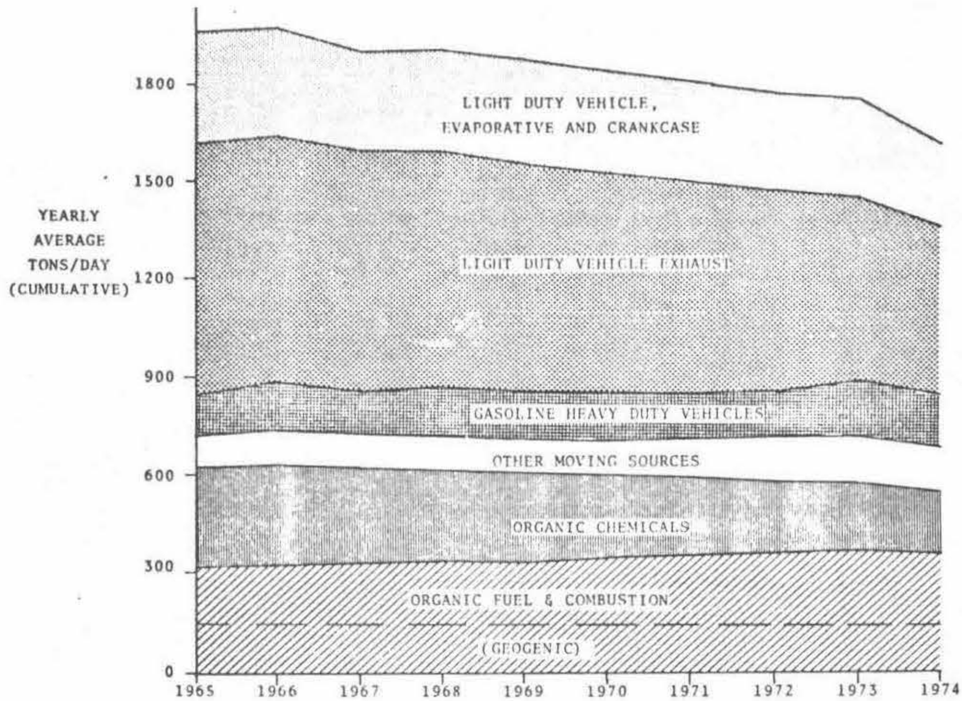

Figure 3. Total Reactive Hydrocarbon Emission Trends in the Los Angeles Region

The net change in basin-wide reactive hydrocarbon emissions over the decade was a decrease of 18 percent. Most of this reduction was due to controls on light-duty vehicles. Light-duty vehicle crankcase emissions were reduced in the early and middle 1960's. Slight reductions in evaporative emissions were obtained in the 1970's from the new car evaporative controls. Exhaust emission standards for new automobiles resulted in significant control of exhaust emissions. The net change in RHC emissions for the average vehicle over the decade was a 52 percent decrease. In the presence of a 41 percent increase in VMT, the net reduction in total RHC emissions from light-duty vehicles was only 32 percent.

Gasoline-powered, heavy-duty vehicles experienced a 23 percent reduction of per-mile RHC emissions over nine years, most of the reduction coming from crankcase emission control. With the 41 percent growth in VMT, the net change was a seven percent increase over the period from heavy-duty vehicles. Emissions from other mobile sources have increased by 56 percent.

The organic chemicals category, the largest contributor of RHC in the stationary source category, has been reduced by approximately 33 percent from 1966 to 1974 due to APCD Rule 66 in the late 1960's and to substitution of water-based coatings for organic solvent coatings in the early 1970's. Emissions from gasoline marketing, which is a major source of organic fuel emissions, have increased by 26 percent, although filling of underground gasoline station tanks has been partially controlled by local APCD rules. An important part of the organic fuels and combustion category is the "geogenic source" that was recently pointed out by Mayrsohn and Crabtree [17]. It is assumed that this source remained constant over the ten-year period.

Geographical Distribution of Emission Trends

The spatial distribution of emissions, as well as the total amount of emissions, is important to ambient air quality. Accordingly, trends in the spatial distribution of emissions should be considered in analyzing air quality trends. This section illustrates large-scale trends in the geographic distribution of emissions within the Los Angeles air basin by documenting net emission changes over the decade on a county-by-county basis.

There is reason to expect that the spatial distribution of emission trends has been very nonuniform in the Los Angeles region. Although similar emission control policies apply to each county, growth rates vary considerably from county to county. Figure 4 illustrates the spatial pattern of VMT and population growth from 1965 to 1974. Growth rates for each individual county are presented within the boundaries of those counties, while basin-wide figures are given in the lower left-hand corner of the figure.

As indicated in Figure 4, the average VMT and population growth rates for the entire basin are 3.9 percent per year and 1.1 percent per year, respectively. Los Angeles County has been growing at a rate considerably slower than the other five counties within the Basin. The fastest growing county, Orange County, has a VMT growth rate (7.5 percent per year) triple that of Los Angeles County (2.8 percent per year) and a population growth

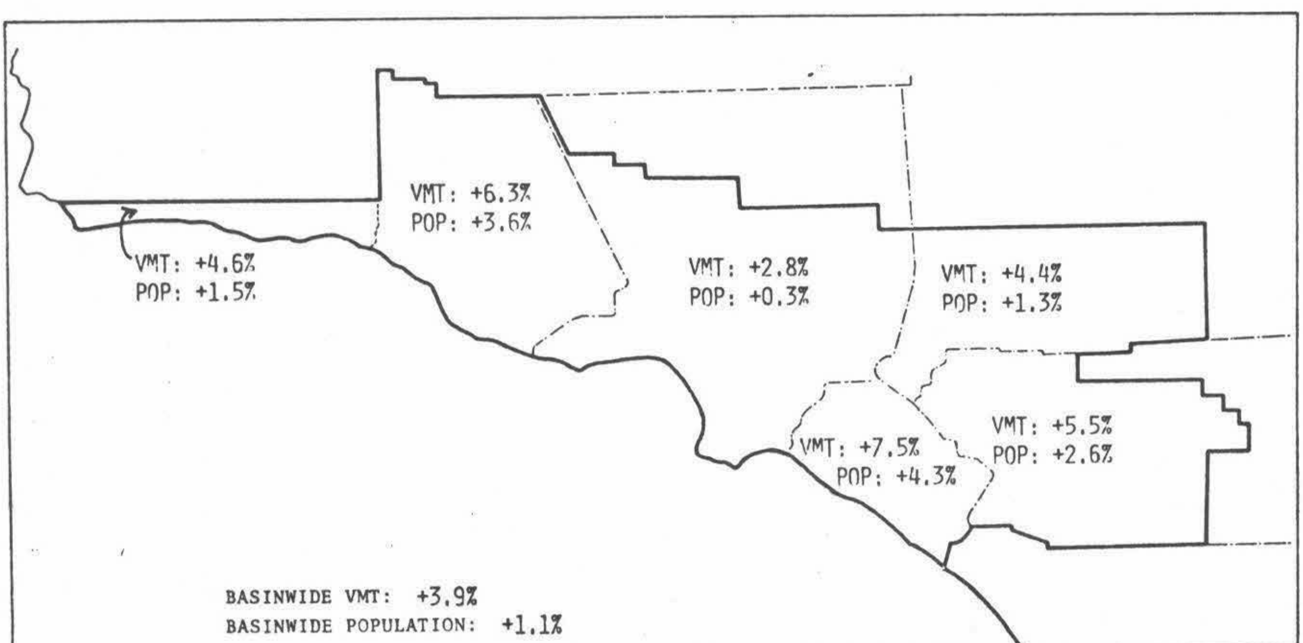

Figure 4. Average Yearly VMT and Population Growth Rates, 1965-1974

rate (4.3 percent per year) more than ten times that of Los Angeles County (0.3 percent per year). These differential growth rates, compounded year by year over the decade, led to a spreading out of emissions away from Los Angeles County to the outlying counties. Los Angeles County accounted for about 75 percent of basin-wide emissions of NO_x and RHC in 1965, but only for about 65 percent in 1974.

Figure 5 illustrates the spatial distribution of NO_x emission changes over the decade. The net percentage change in NO_x emissions over the decade is presented for each of the six counties and for the entire basin. Each county experienced an increase in NO_x emissions over the decade, with the basin-wide total increasing by 36 percent. Following the spatial trends in population and VMT growth, Los Angeles County NO_x emissions grew by the smallest amount, 25 percent. NO_x emissions in Orange County rose drastically, an estimated 89 percent increase over the decade.

Figure 6 presents the county-by-county distribution of RHC emission changes. Over the decade, basin-wide RHC emissions decreased by 18 percent. Following the order of population and VMT growth rates, Los Angeles County showed the largest decrease, 24 percent, while the outlying counties experienced lesser decreases. In fact, Orange County evidently underwent an actual increase in RHC emissions of six percent over the decade.

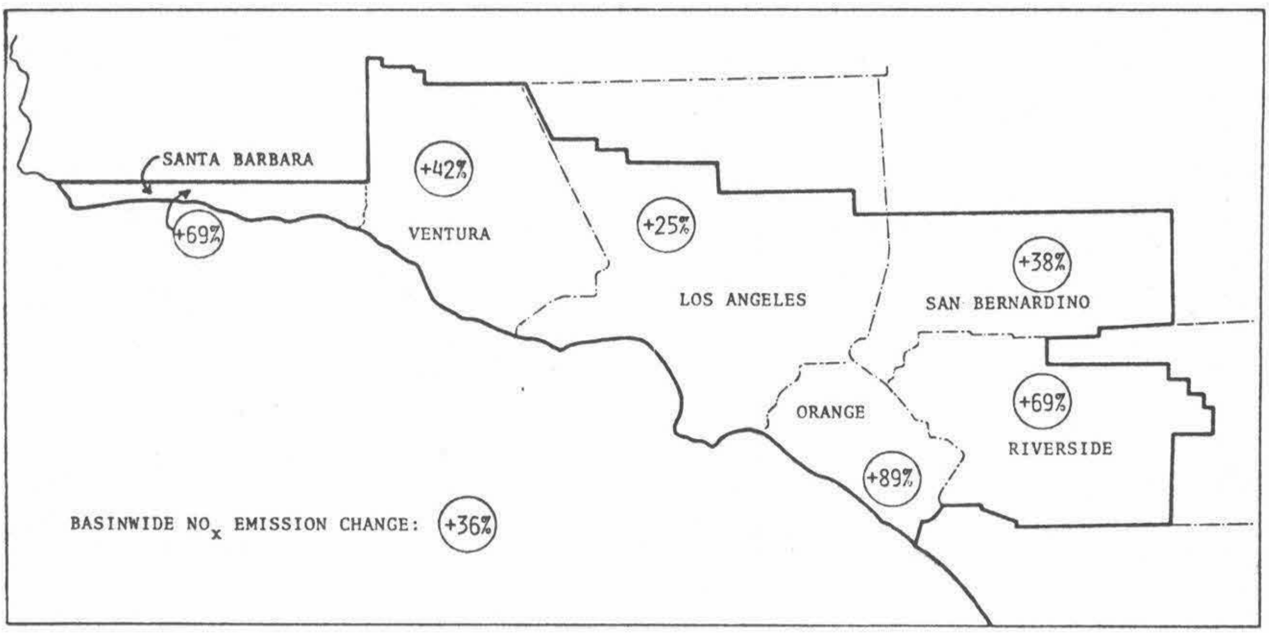

Figure 5. Trends in NO_x Emissions, 1965-1974

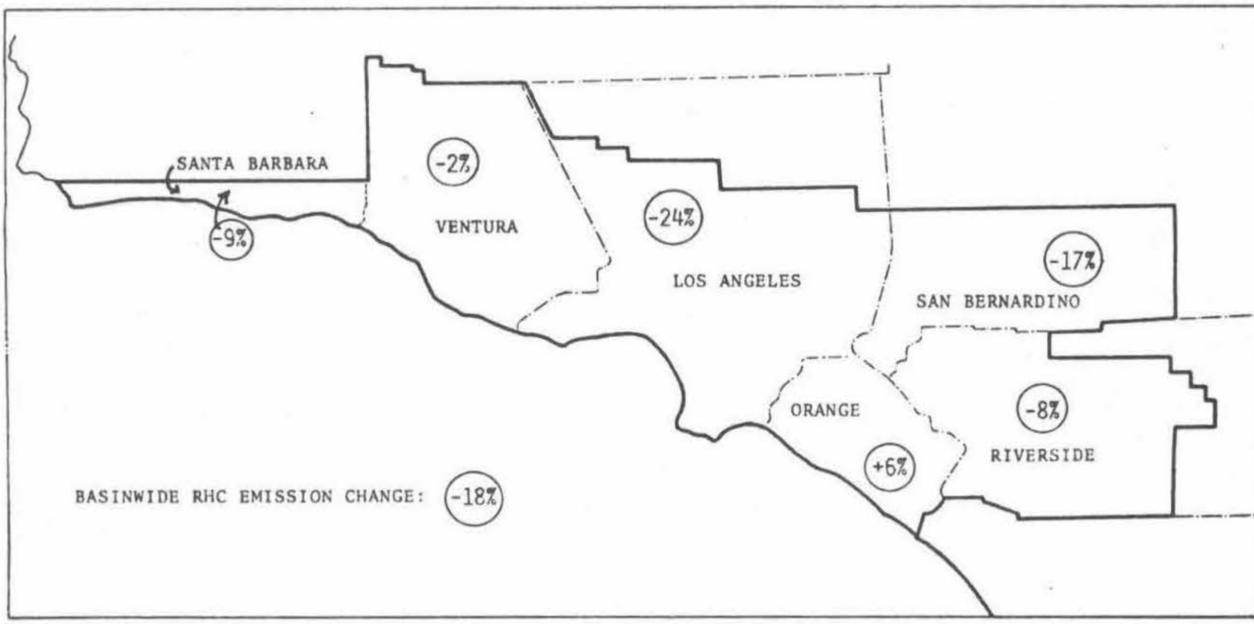

Figure 6. Trends in RHC Emissions, 1965-1974

3. AMBIENT AIR QUALITY TRENDS

Having documented emission trends in the Los Angeles region over the past decade, the next logical step is to compare the emission trends to air quality trends. This section presents air quality trends over the decade for both oxidant and its precursors and analyzes these trends in terms of the changes in total emissions and the spatial distribution of emissions. Comparing ambient precursor trends to emission trends provides a test of the adequacy of simple proportional (rollback) models for relating emissions and precursor concentrations. This juxtaposition also serves as a partial check on both the emission and air quality data bases. Comparing oxidant trends to precursor trends yields insight as to the oxidant/precursor relationship in various parts of the Los Angeles region.

The data base for determining air quality trends in the Los Angeles region consisted of computer tapes of hourly average pollutant measurements obtained from the California Air Resources Board. These tapes contained data from all stations in the Los Angeles region which reported to the Air Resources Board during the years 1963 to 1974. A search was conducted among personnel of the ARB and the County APCD's to determine whether monitoring changes occurred which could affect air quality trends. Data were included from each site only if they were taken with the same measurement method each year. It was determined that the locations of monitors in a few cities had been altered during the period of interest; data from these locations were eliminated from the study. Some of the monitoring sites that were included in the trend analysis are shown in Figure 1.

In order to compare air quality changes to emission changes over the decade, net changes in ambient concentrations were determined from 1965 to 1974. For each pollutant (NO_x, NMHC, and oxidant), a least-squares trend line was fit to the yearly averages of daily maximum one-hour concentrations. The percentage change in this trend line from 1965 to 1974 was used as the measure of overall air quality change for each site. Yearly averages of daily maxima were eliminated from the analysis if the averages were based on a very limited number of sampling days. The analysis was performed only for stations with at least eight years of adequate data.

Ambient Precursor Trends

Figure 7 compares changes in NO_x air quality to changes in NO_x emissions. The circled numbers in Figure 7 represent the net percentage emission changes for each county over the decade (also given in Figure 5). For instance, Los Angeles County experienced a 25 percent increase in NO_x emissions over the decade, Orange County experienced an 89 percent increase, Riverside underwent a 69 percent increase, etc. The net percentage changes in ambient NO_x concentrations over the decade are also plotted in Figure 7 for eleven monitoring sites. For instance, ambient NO_x levels increased by 8 percent at downdown Los Angeles, 76 percent at Anaheim, 47 percent at San Bernardino, etc. Basin-wide averages in emission trends and ambient concentration trends are presented in the lower left-hand corner of Figure 7.

The agreement between calculated NO_x emissions changes and measured NO_x air quality changes is strikingly good. Total basin-wide NO_x emissions in creased by 36 percent over the decade, while average NO_x concentrations at

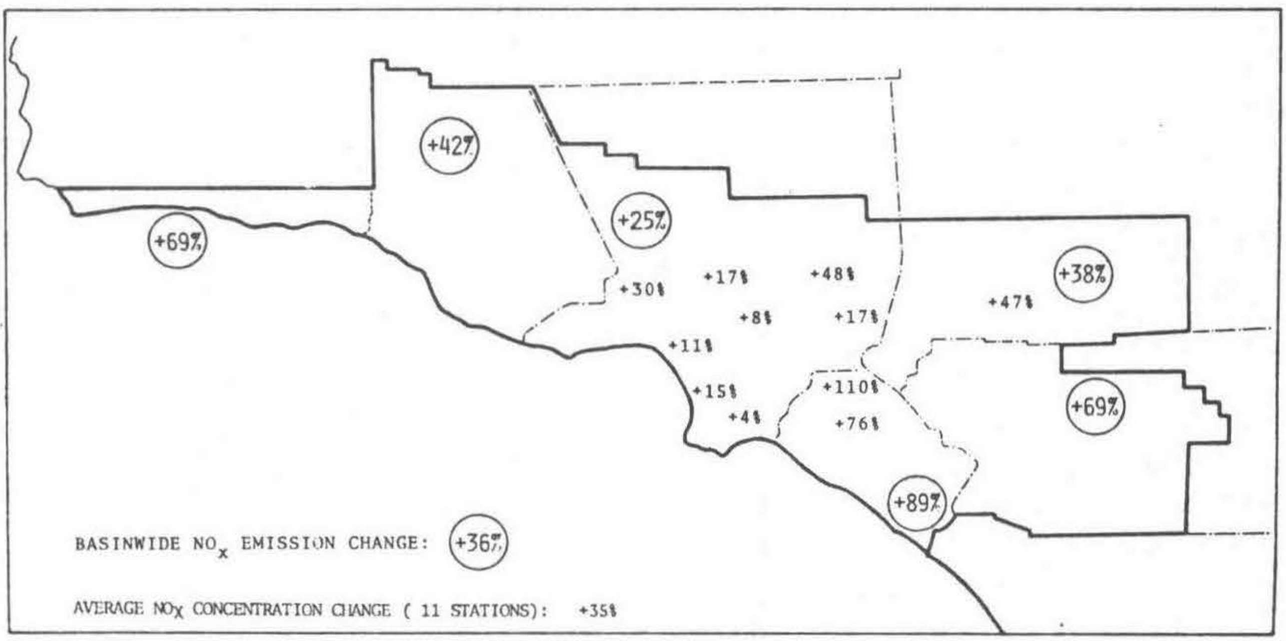

Figure 7. Trends in NO_x Emissions and Air Quality, 1965-1974

the eleven monitoring sites increased by 35 percent. Even the spatial distribution of emissions and air quality trends agree on a county-by-county basis. NO_x emissions grew by 89 percent in Orange County, while the two Orange County stations showed an ambient NO_x increase of 93 percent. NO_x emissions increased by 25 percent in Los Angeles County, while the eight Los Angeles County stations experienced a 19 percent increase in measured NO_x concentrations.

Figure 8 compares changes in nonmethane hydrocarbon (NMHC) concentrations to changes in RHC emissions. The circled numbers again represent net percentage emission changes, for each county and for the entire basin. Only four monitoring sites provide data sufficient for determining ambient hydrocarbon trends. The net percentage changes in NMHC* levels over the decade are plotted at the locations of these four sites.

It is not possible to draw definitive conclusions concerning ambient NMHC trends because of the sparsity of sites providing data on changes in hydrocarbon concentrations. However, it is encouraging that the average change in ambient NMHC concentrations among the four sites, a 15% decrease, is close to the basinwide change in RHC emissions, an 18% decrease. Also, the spatial pattern of concentration changes seems to agree qualitatively with the spatial pattern of emission changes (at least in the sense that Los Angeles, San Bernardino, and Orange counties fall in the same order for concentration trends and emission trends). The only puzzling aspect of the ambient trend data for NMHC is the wide disparity between trends at the two sites in Los Angeles County, Downtown Los Angeles and Azusa.

*Nonmethane hydrocarbon trends are computed from total hydrocarbon trends using empirical formulas relating THC to NMHC [18].

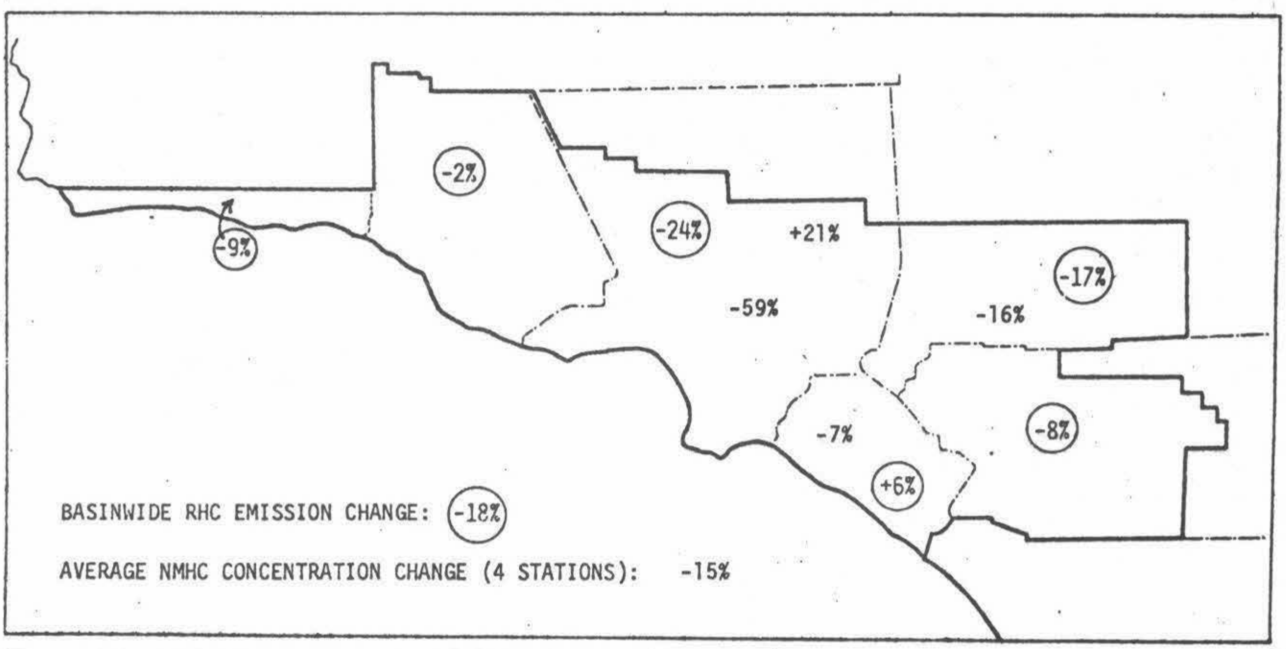

Figure 8. Trends in RHC Emissions and Air Quality, 1965-1974

Trends in Oxidant Air Quality

Figure 9 illustrates percentage changes in oxidant concentrations over the past decade and compares these to changes in RHC emissions. The circled numbers represent RHC emission trends (county by county and Basin-wide) while the other numbers represent trends in ambient oxidant levels at thirteen monitoring sites. Basin-wide, the agreement between RHC emission trends and oxidant trends is quite good; total RHC emissions decreased by 18 percent over the decade, while the average improvement in oxidant levels at thirteen stations was 19 percent. Also, the spatial distribution of air quality changes agrees qualitatively with the spatial distribution of emission changes, at least in the sense that Los Angeles County experienced the greatest improvement in both RHC emissions and oxidant concentrations.

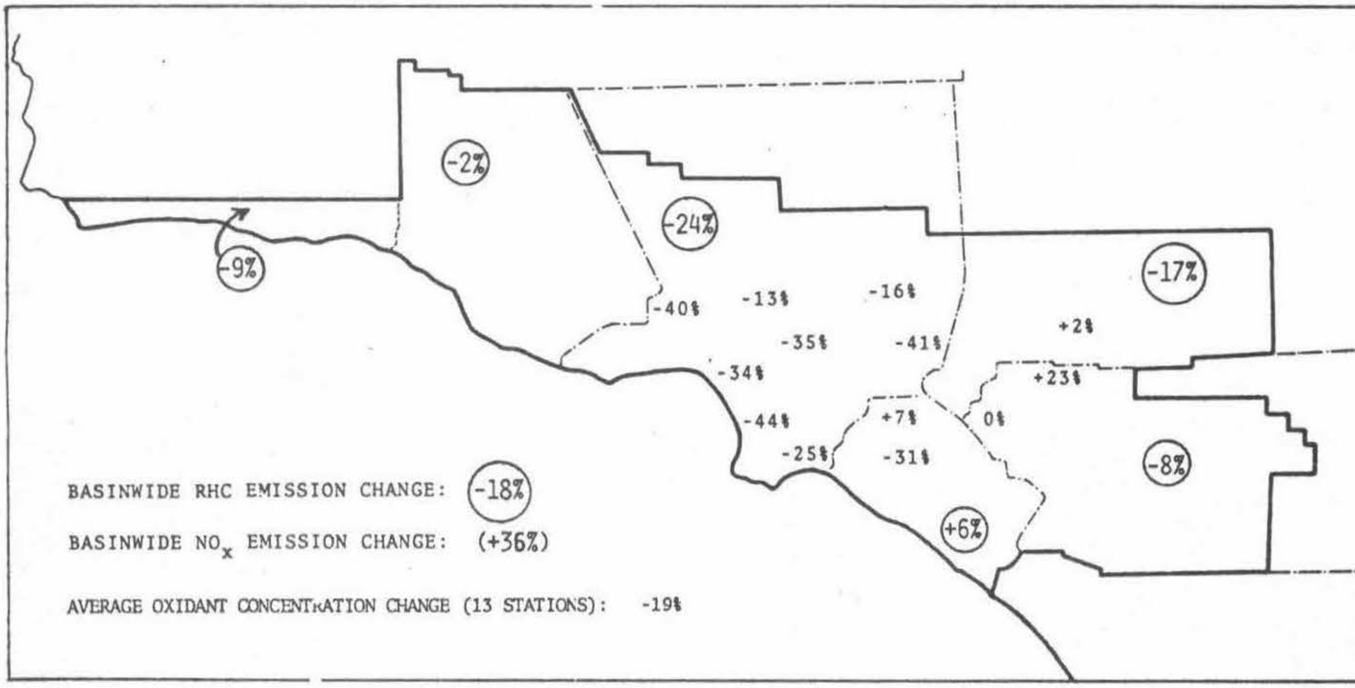

Figure 9. Trends in RHC Emissions and Oxidant Air Quality, 1965-1974

The spatial pattern of oxidant air quality trends can be even better explained by considering what has been termed the "dual role" of NO_x emissions in oxidant formation. From the present understanding of photochemistry, it can be argued that, at the existing ratio of RHC to NO_x, increases in NO_x emissions would tend to decrease oxidant concentrations in the source-intensive areas of the Basin (the central and western-coastal areas). In these areas, the highest oxidant concentrations occur early in the day (around noon) before the pollution is transported inland by the daily sea breeze. The most effective strategy for reducing oxidant near the source areas early in the day is to inhibit the reaction by reducing the RHC/NO_x

ratio. This can be done by decreasing RHC emissions and/or increasing NO_x emissions. The transported oxidant problem in the eastern-inland areas should depend more on the absolute levels of RHC and NO_x than on the ratio. The key to controlling the transport problem is less understood, but indications are that reductions are needed in overall levels of RHC and (possibly) NO_x.

These observations help to explain why the coastal areas of Los Angeles County show the greatest improvement in oxidant concentrations (a 30 to 40 percent decrease from 1965 to 1974). These source-intensive areas have experienced a 24 percent decrease in RHC emissions, and they have been aided by the NO_x emission increase. The apparent anamoly at Anaheim, a 31 percent decrease in oxidant within a county with a 6 percent increase in RHC emissions, can be partially accounted for by the large NO_x emission increase (and resulting RHC/NO_x ratio decrease) at that source-intensive location. The lesser rates of improvement in the eastern-inland portions of the basin may be explained by noting that basin-wide RHC emissions have been reduced only slightly and that total NO_x emissions have increased. In particular, Riverside may have experienced a 23 percent increase in oxidant concentrations because it is downwind of Orange County, which had a 6 percent increase in RHC emissions and a large increase (89 percent) in NO_x emissions.

4. CONCLUSIONS

Basin-wide emissions of NO_x have increased by 36% in the Los Angeles region from 1965 to 1974, while basin-wide RHC emissions have decreased by 18%. Controls on crankcase and exhaust emissions from light-duty vehicles were largely responsible for the total RHC emission decrease. VMT growth and the increase in NO_x exhaust emissions from 1966 to 1969 model year vehicles were the major contributors to the total NO_x increase.

Projections of emissions made in the early 1970's [19] have turned out to be overly optimistic when compared to emission trends which we now determine in retrospect. The projections made in the early 1970's indicated a 20% increase in NO_x emissions and a 40% decrease in RHC emissions from 1965 to 1974. The error in the earlier projections is caused partly by the fact that test data for automobiles do not show as large emission reductions as would have been expected from the time-table of tightening emission standards. Evaporative emissions, for example, are much higher than called for by federal and state standards. For reactive hydrocarbons, the new reactivity scale we are using now leads to a more pessimistic picture because it gives greater weight to source categories that have been reduced little in the last ten years. For NO_x, the shortage of natural gas for industrial interruptibles and power plants and the resultant unforeseen switch to fuel oil have produced higher emissions than originally projected for stationary sources. The occurrences of these unforeseen circumstances should serve as a warning in making future emission projections. A sensitivity analysis of air quality to the possible range of emissions should be included as part of future studies.

Emission trends reflect a competition between reductions from emission control and increases from source growth. Since the geographical distribution of source growth, in particular traffic growth, has been very nonuniform over the Los Angeles region, the geographical distribution of emission trends is also very nonuniform. Among the counties in the basin, Los Angeles County has grown the slowest, while Orange County has expanded at the greatest rate. This nonuniform spatial distribution of emission trends is important in interpreting air quality trends. What appear to be contradictory air quality trends at different locations in the basin can often be explained by the geographical distribution of emission trends. In analyzing future strategies, the geographical distribution of emission changes should be accounted for.

The basin-wide trend in ambient NO_x concentrations (about a 35 percent increase) agrees well with the basin-wide trend in NO_x emissions over the past decade (a 36 percent increase). The geographical distribution of NO_x emission trends and NO_x air quality trends on a county-by-county basis also agree. Ambient data on hydrocarbon trends are very sparse. However, the average trend in NMHC concentrations at four locations (a 15 percent decrease) is consistent with the basin-wide change in RHC emissions (an 18% decrease).

The basin-wide improvement in oxidant air quality (19 percent over the decade) agrees well with the basin-wide reductions in RHC emissions. The geographical trends in oxidant agree, qualitatively, with the geographical trends in RHC emissions, especially for Los Angeles County. If we account for the dual role of NO_x we obtain an even better clarification of the geographical distribution of oxidant trends. The increase in NO_x emissions helped to decrease the RHC/NO_x ratio; this resulted in substantial oxidant reductions in the source-intensive, central-coastal areas. Little or no improvement in transport related oxidant occurred in the downwind, eastern-inland areas because overall RHC emissions were reduced only slightly and (possibly) because NO_x emissions increased.

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