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The Ozone Weekend Effect in California: Evidence Supporting NO_x Emission Reductions

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ABSTRACT

Ozone is typically higher on weekends (WE) than on weekdays (WD) at many of California's air-monitoring stations. Sometimes called the "ozone WE effect," this phenomenon occurs despite substantially lower estimates of WE emissions for the major ozone precursors – volatile organic compounds (VOC) and oxides of nitrogen (NO_x).

Compared to WD emissions, WE emissions of NO_x decrease more (proportionally) than do the WE emissions of VOC. Because the WE increases in ozone coincide with the relatively large WE reductions in NO_x, some conclude that regulations that would reduce NO_x emissions on all days would undermine ozone attainment efforts by causing ozone to decrease more slowly (or even to increase).

At this time, public discussion of the ozone WE effect has mostly reflected the viewpoint that NO_x emission reductions would not help reduce ambient ozone levels. A large body of published research from this perspective has accumulated over the last 10 to 20 years. Nevertheless, the presently available scientific evidence can also lead to the conclusion that NO_x emission reductions may be needed to maintain or even to expedite progress toward attainment of ozone standards. New field studies, selected laboratory experiments, and improved models would be needed to resolve the central issues and help determine the best interpretation of the evidence.

The evidence for the viewpoint that NO_x emission reductions may be needed to maintain or even to expedite progress toward attainment of ozone standards comes from widely scattered sources and has only recently been assembled in an integrated fashion. This paper emphasizes scientific assessments of the available data and provides a systematic interpretation of them. In this regard, the following conclusions and observations are important:

- The ozone WE effect is not, as some have suggested, a "real-world" test of the ozone air quality impact of California's NO_X control program. The spectrum of differences that occur from WD to WE includes some potentially important factors beyond those emissions changes and impacts commonly associated with control programs.

  Regulations that reduce NO_x emissions typically achieve these reductions on a steady and consistent basis. The WE reductions of NO_x and VOC, on the other hand, occur on a seven-day cycle that temporarily interrupts the greater production of emissions characteristic of weekdays. Changes in the composition (including the amount), timing, and location of emissions on WE vs. WD alter the ozone-forming system in ways that do not mimic its response to the steady changes that would result from the reductions in NO_x emission planned for California in the coming decade(s).

  For 20 years or more, WD ozone levels decreased faster than did WE ozone levels, and WE have now been left behind. At the same time, VOC emissions were reduced substantially faster (by 50% or more) compared to NOx emissions. The associations between these emission reductions and the slower WE response justify the following important conclusion -- continued emphasis on VOC reductions may undermine attainment goals by allowing the

highest ozone days, which now occur on weekends, to lag behind as progress continues to be fastest on weekdays.

- At peak ozone sites (sometimes called "design sites"), the ozone WE effect is usually no more than half of that found at some other locations in a planning area. Ozone concentrations at design sites determine whether a region attains the federal and state ozone standards. Design sites tend to be in the less urbanized areas "downwind" of major emission source regions, and reductions of NO_x emissions are often expected to reduce ozone peaks in such areas. A complete picture of the cause(s) of the ozone WE effect may reveal that NO_x reductions are needed to expedite attainment of ozone standards at design sites.

- On the worst ozone days of the year, the very days that determine attainment status, the ozone WE effect appears to be relatively small. That is, the ozone WE effect seems to be smallest when the ozone-forming potential is greatest. An important meteorological condition that leads to high ozone-forming potential is stagnation caused by high pressure systems that encourage pollutants to build up over two or more days. During such multi-day episodes, VOC concentrations increase substantially more than do NO_x concentrations. Under such circumstances, NO_x may be the limiting factor in ozone formation. Therefore, NO_x reductions may have their greatest ozone-reducing benefits on those days that determine whether regions attain ozone standards.

- Most of the ozone that forms from VOC and NO_x emissions is not generated at ground level; rather it forms aloft in a layer of air between 100 and 1500 meters above the surface. Pollutant measurements within this layer aloft are scarce, but the available evidence suggests that ozone formation may be limited by the availability of NO_x rather than by the availability of VOC. If so, NO_x reductions should reduce the amount of ozone that forms aloft. This would be especially true during multi-day episodes when VOC accumulates more than NO_x and ozone concentrations reach their highest levels.

- Ozone that forms aloft and mixes down toward the surface does not always survive long enough to be measured by routine monitors on the ground. The main reason is that fresh NO_x emissions destroy ozone before these monitors can measure it. Freshly emitted NO_x includes NO, which destroys ozone through a fast reaction referred to colloquially as "scavenging" or "quenching". In this reaction, NO takes an oxygen atom from ozone (O_3) to produce NO_2 and common oxygen (O_2). Emissions of NO_x are mostly NO (90% or more), and most NO_x is emitted at or near ground level. On weekends, little NO is emitted compared to weekdays, particularly during the daylight hours when ozone is formed and vertical mixing is typically most active. Though not definitive, the limited data on conditions aloft indicate that ozone concentrations aloft may be routinely higher on weekdays than on weekends, but less of this ozone reaches the surface on weekdays than on weekends due to surface-level quenching by fresh NO_x emissions.

- Large reservoirs of ozone and ozone precursors often carryover aloft from one day to the next. This ozone, along with ozone that forms aloft the same day, eventually mixes down toward the surface. Because NO_x emission reductions are likely to reduce ozone levels in reservoirs aloft, NO_x reductions should reduce the potential (see previous bullet) impact of ozone carryover aloft on ozone measured at the surface. The ozone-reducing potential of NO_x reductions would be especially important on WE when lower NO_x emissions at the surface permit ozone to mix down from aloft and be measured by ground-level monitors.

- Photochemical simulation models continually increase in sophistication and realism. Nevertheless, the procedures and databases on which they rely can never capture every detail of the atmospheric chemistry and physics in the real world. Models must rely on assumptions that simplify both the data and the calculations required.

  The models and databases available today have helped to clarify issues and inform decisions. For investigating the cause(s) of the ozone WE effect, however, today's models are lacking in some critical areas. Issues regarding the completeness of weekend inventories and chemical mechanisms are subjects of ongoing research and development. Additional concerns involve the realism of processes that simulate horizontal and vertical transport and the appropriateness of the vertical layers used to represent the air above the ground. These issues may also be resolved through new or ongoing research. At this time, however, we do not consider the models available to be adequate tools for determining the cause(s) of the ozone WE effect and for quantifying their respective contributions.

Taken together, the observations above show that NO_x emission reductions may be needed to expedite overall progress toward the goal of attaining ozone standards. Clearly, it is premature (at least) to conclude that the ozone WE effect demonstrates that NO_x emission reductions would delay attainment of ozone standards.

INTRODUCTION

Ozone concentrations measured on weekends (WE) are typically higher than those measured on weekdays (WD) in many parts of California.[1] This phenomenon, referred to here as the "ozone WE effect," occurs despite substantially lower WE emissions estimates for the main ozone precursors – volatile organic compounds (VOC) and oxides of nitrogen (NO_x). Although the ozone WE effect has been studied for more than twenty years,[2,3,4] a validated explanation for it is not yet available.

The issue is controversial because the ozone WE effect is viewed by some as proof that NO_x-reducing regulations are unwise because they would undermine the progress that VOC reductions alone would otherwise achieve. If this view is correct, so-called "NO_x disbenefits" would delay or even prevent attainment of ozone air quality standards.

This paper presents an alternative explanation of field observations and model results. According to this explanation of the evidence, NO_x reductions not only would not jeopardize ozone attainment goals but would further these goals expeditiously.

ANALYSES CONCERNING THE OZONE WEEKEND EFFECT

To help organize our thoughts concerning the ozone WE effect and its implications, it may be helpful to keep the following questions in mind:

1. When ozone molecules are detected at a surface monitor,
   - Where were they generated?
   - When were they generated?
   - Under what conditions were they generated?
2. How do the answers to these questions differ on weekdays versus weekends?
3. What do these differences imply regarding responses to regulatory (steady) reductions of VOC and NO_x rather than weekend (intermittent) reductions of VOC and NO_x?

Though complete answers to these questions are not available at this time, the analyses presented in this paper provide helpful input for their consideration.

Ozone and Emission Trends

In California, historical changes in ozone in response to thirty years of VOC and NO_x reductions lead to an inescapable conclusion: while VOC emissions decreased faster (by 50% or more) than NO_x emissions, ozone decreased faster on weekdays than on weekends. Ozone in some areas decreased strongly on both weekdays and weekends, but comparatively, weekends were "left behind." Two analyses illustrate the patterns that lead to this conclusion.

In the first analysis, we compared ozone trends for weekdays and weekends from 1980 to 1998 in the South Coast Air Basin (SoCAB).[5] These nineteen-year trends illustrate the patterns clearly and avoid some arcane but important issues that arise when combining data from the early 1970's. For each year, daily maximum ozone measurements were summarized by day-of-week.

For each day of the week, the day with the highest ozone measurement was discarded and the ten highest remaining values were averaged to represent the year. The ten values selected represent the highest one third of the values during the ozone season for each day of the week. Figure 1 shows the WD and WE trends for ozone at five sites in the SoCAB. Arranged from west to east, the sites are Los Angeles, Azusa, Upland, Riverside, and Crestline (Lake Gregory). This sequence also represents increasing distance from emission sources in the central urban region

The trends in Figure 1 clearly illustrate how ozone on weekends has improved more slowly compared to ozone on weekdays, and that WE ozone now surpasses WD ozone at each site. Today, the highest ozone concentrations in the South Coast Air Basin (SoCAB) typically occur on weekends, especially on Sunday, rather than on weekdays. Sundays also tend to record the highest ozone concentrations in the San Francisco Bay Area Air Basin (SFBAAB), the San Joaquin Valley Air Basin (SJVAB), and the Sacramento Metropolitan Area (SMA), though the differences with respect to weekdays are much less pronounced in the latter two regions (see Table 3).

The transition of high ozone days from weekdays to weekends occurred while VOC emission reductions outpaced NO_x emission reductions. Table 1 presents emissions inventories[6] of VOC and NO_x for 1980 and 2000 in four areas of California, including the SoCAB, where the rate of VOC reductions has been about 50% faster than the rate of NO_x reductions. Table 2 shows the percent decrease in ozone on weekdays and weekends over this period for five separate sub-regions of the SoCAB. Although ozone decreased on all days of the week in all five sub-regions, ozone on weekdays improved significantly more than it did on weekends.

In a second analysis, Tran and Austin[1] prepared a rigorous assessment of the ozone WE effect in the SoCAB, the SFBAAB, and the SMA. The methods they applied included appropriate filters to remove long-term and seasonal trends, an explicit accounting for serial dependency, and robust estimation of means to limit the impact of outliers. Their work considered the ozone WE effect before and after the introduction of California's Phase 2 reformulated gasoline (CaRFG2), which occurred in 1996. According to emissions inventories, CaRFG2 reduced total VOC emissions about 50% more than it reduced NO_x emissions.[7] Although CaRFG2 has been credited with reducing ambient ozone generally,[7] the gap between weekdays and weekends widened significantly in all three air basins immediately following the introduction of CaRFG2. In addition, Sunday emerged as the day-of-week with the highest ozone concentrations after the introduction of CaRFG2 in all three areas considered by Tran and Austin.[1]

An important conclusion is justified by these two analyses: continued emphasis on VOC reductions may undermine attainment goals by allowing the highest ozone days, which now occur on weekends, to lag behind as progress continues to be greatest on weekdays.

Since an emphasis on VOC reductions has left weekends behind, some obvious questions arise. Should NO_x reductions proceed at the rate of VOC reductions in order to reduce the highest ozone concentrations in California, which now occur on weekends, on Sundays in particular? When ozone molecules are measured at the surface on Sundays, where were they generated and under what conditions? These thoughts are explored is systematic fashion in the remainder of this paper.

Spatial Distribution of the Ozone Weekend Effect

In California air basins, the ozone WE effect is relatively small in the areas that determine attainment status. These are the most relevant areas when considering possible benefits or disbenefits of NO_x emissions reductions with respect to the goal of attaining ozone standards.

Previous research quantified the ozone WE effect at monitoring sites in four major areas of California. In addition to the three air basins considered by Tran and Austin,[1] the staff of the California Air Resources Board (CARB) quantified the ozone WE effect in the San Joaquin Valley Air Basin.[8] As shown in Table 3, the ozone WE effect in all four regions is significantly greater (proportionally) in the urbanized areas than in the suburban and rural areas downwind. Table 4 identifies the monitoring locations (by AIRS identification number) that were considered in each category. The suburban and rural areas that exhibit smaller WE effects are the same areas that record the highest ozone concentrations in each region. In other words, smaller WE effects usually occur in those areas that determine a region's attainment status. In these areas, the ozone WE effect (averaged over the whole ozone season) is about 20% in the South Coast and San Francisco Bay Area Air Basins and 5% in California's interior valleys.

This spatial pattern in the ozone WE effect may occur for at least two reasons that are not mutually exclusive; both may play a role in determining the overall WE effect. The first reason relates to classic results in photochemistry in which ozone formation in areas near emission sources tends to be VOC-limited while ozone formation moves from VOC-limited to NO_x-limited as the distance downwind from emission sources increases. Those who view the WE effect as a demonstration of "disbenefits" due to lower NO_x and higher VOC/NOx ratios on weekends discuss this viewpoint in detail. With few exceptions, the published literature on the ozone WE effect takes this point of view.

The second reason for the spatial pattern in the ozone WE effect relates to another well-known phenomenon in ozone photochemistry – "scavenging" or "quenching" of ozone molecules by nitric oxide (NO). The role that scavenging of ozone by NO appears to play in the ozone WE effect is discussed at some length in the next section.

The Role of Ozone Scavenging by NO in the Ozone Weekend Effect

Although NO_x is composed of both NO and NO_2, the primary species in fresh NO_x emissions is NO, typically 90% or more.[9] The NO emissions scavenge ozone according to a fast reaction in the atmosphere. Ozone (O_3) will combine with NO to produce O_2 and NO_2. The prevalence of this reaction appears to play a major role in the ozone WE effect.

Emissions of NO at the surface in urbanized areas are relatively abundant on weekdays but relatively scarce on weekends, particularly on Sundays. Fresh NO emissions at ground level quickly scavenge ozone molecules and reduce the ozone concentrations measured by surface monitors. This scavenging phenomenon is much greater on weekdays than on weekends. Four observations are given to support this claim.

Day-of-week profiles for traffic and NO_x. As part of a larger investigation of the ozone WE effect, we analyzed patterns in traffic data and compared them to patterns in air quality data. These analyses were limited to the SoCAB, but they yielded significant results that may represent other urbanized areas.[10,11]

Motor vehicle activity differs dramatically between weekdays and weekends, especially Sundays. The largest discrepancy between weekdays and weekends is between 6:00 a.m. and 11:00 a.m., with lesser but possibly important differences between 3 p.m. and 7 p.m. Figure 2 shows day-of-week composite profiles for hourly volumes of light-duty vehicles at 15 Weigh-in-Motion (WIM) stations in the SoCAB. Figure 3 shows these patterns for heavy-duty vehicles for the same stations. The figures are based on data archived by the California Department of Transportation throughout the year 2000. These figures are similar to those in the work cited but represent a full year of more recent data.

Figure 4 shows composite profiles for day-of-week NO_x measurements at eleven air quality monitors in Los Angeles and Orange Counties, many of them near the WIM stations used to characterize traffic patterns. The profiles for traffic and for NO_x are consistent with emission inventory data[11] for the year 2000 that indicate over 60% of NO_x emissions in the SoCAB are produced by on-road motor vehicles. When all mobile sources are considered, they account for almost 90% of the NO_x emissions in the SoCAB. Source tests show that NO_x emissions are predominantly in the form of NO, which is converted to NO_2 and other nitrogen-containing species in the atmosphere.[9]

From the air quality and emissions inventory data, it is clear that fresh NO emissions are much greater on weekdays than on weekends, especially on Sundays. The opportunity for fresh NO emissions to scavenge ozone near the surface is much greater on weekdays than on weekends.

Vertical profiles for NO_x and ozone. Vertical profiles for NO_x and ozone are not commonly available apart from intensive field studies. Major field studies of ozone in Southern California were conducted in 1987 and 1997. Both studies included some vertical profiles of ozone and/or NO_x measured by aircraft, ozonesondes, or lidar. The studies, however, were not designed with the WE effect in mind. In fact, the 1987 study sought representative weekday episodes, while the 1997 study sought representative WE episodes. Ambient ozone concentrations and emissions of ozone precursors both declined dramatically in the ten years between these studies. As a result, weekdays and weekends between and within the studies are not easily compared with confidence.

An analysis of the ozonesonde data from the 1997 Southern California Ozone Study (SCOS97) suggests that afternoon differences between ozone at the surface and at 100 meters are greater on weekdays than on weekends. For this analysis, we estimated the ozone concentration at 100 meters by interpolating between the nearest values below and above 100 meters. For each day, the measurements closest to the surface after equilibration were used to estimate the surface concentration. Because a measure of subjective judgement was required in this analysis, we took care to err toward equal ozone values at the surface and aloft. The results, shown in Table 5, may indicate that ozone on weekdays is prevented from mixing down to ground level where surface monitors can measure it. If so, the prime suspect is an extra measure of scavenging by fresh NO emissions near the surface on weekdays that does not occur on weekends.[1]

Different WE effects at two nearby locations. The highest ozone concentrations in the SFBAAB are typically measured at Livermore. Recently the Livermore monitor was moved about 1.5 km. The move was precipitated by analyses showing that NO emissions from a newly activated bus terminal a short distance upwind of the old location were scavenging ozone and depressing the measured ozone concentrations. Like most public transit systems, activity at the bus terminal was much lower on weekends than on weekdays. Route schedules from the Livermore Amador Valley Transit Authority show greatly reduced service on Saturdays and no service at all on Sundays.

At the old monitoring location, daily maximum ozone concentrations on Sundays averaged about 32 percent higher compared to Fridays. For 2000 and 2001, the first two ozone seasons at the new monitoring location, the daily maximum ozone on Sundays averaged only 17 percent higher compared to Fridays. This was not due to a decrease in ozone on Sundays; rather, the emissions from the bus terminal were no longer quenching ozone on weekdays, so ozone measurements on Fridays increased relative to those on Sundays.

This analysis shows that a significant portion of the ozone WE effect can be due to day-of-week differences in ozone scavenging at the surface by fresh NO emissions. It is possible that much of the remaining 17 percent difference between Sundays and Fridays at the new Livermore location is due to ground level suppression of ozone due to scavenging by NO emissions from on-road and off-road motor vehicles that are plentiful on weekdays but relatively scarce on weekends.

Comparison of total oxidant on weekdays and weekends. If surface emissions of NO depress surface ozone concentrations through scavenging, the WE effect for the sum of ozone and NO_2 (total oxidant) should be smaller than the ozone WE effect. Table 6 summarizes the WE effect for total oxidant in the SoCAB based on the ozone seasons (May through October) for 1998 through 2000. For all locations, the largest WE effect for total oxidant was 9% and the basinwide average was 5%. These are drastically smaller than the values in the SoCAB for the ozone WE effect, for which the largest was 43% and the average was 20%.

If differences in ozone scavenging at the surface on weekdays versus weekends is an important contributor to the ozone WE effect, could there be circumstances in which ozone is so abundant that the scavenging of ozone by NO emissions on weekdays makes less of a difference? The next section considers this question.

[1] The measurement method used with the ozonesondes contributes further bias that minimizes the weekday vs. weekend difference due to scavenging by NO. The "ozone" measurements are based on the KI (potassium iodide) method, which also responds in part to NO_2 and other oxidants. Analyses in Chapter 5.3 of the Technical Support Document show that weekday NO_2 levels are significantly greater than are the weekend levels. Therefore, the weekday vs. weekend contrasts between ozone at the surface and aloft are actually greater than those shown in Table 6.

Ozone-Forming Potential and the Ozone Weekend Effect

We evaluated the ozone WE effect in the SoCAB under varying levels of ozone-forming potential (OFP).[12] The measured ozone levels were not used to characterize OFP because that would have introduced a selection bias. For example, if high ozone on Fridays were the basis for selection, the WE effect would be greatly underestimated because the highest Fridays would not necessarily be paired with the highest Sundays. Selection of Sundays with high ozone would bias the estimation the other direction. Other methods, such as using the weekly average will reduce but not remove selection biases.

To minimize the potential for selection bias, a method of characterizing OFP based on meteorology alone without regard to the measured ozone was used. An equation developed by Larsen[7] in a study of ozone-reducing benefits of reformulated gasoline was used to characterize daily OFP. The equation relates maximum ozone in the SoCAB each day to same-day meteorological measurements that relate to atmospheric dispersion and solar intensity. When values for these meteorological parameters are entered into the equation, they approximate daily OFP as it occurred in the years used to calibrate the equation (1993 and 1994). Measured ozone concentrations have since decreased, but the OFP values from the equation can still be used to indicate relative OFP today.

Using the OFP equation, days between mid-May and mid-October for six years (1992-1994 and 1996-1998) were assigned to three groups. Low OFP was defined as up to 0.12 ppm, medium OFP was between 0.12 and 0.16 ppm, and high OFP was greater than 0.16 ppm. The resulting groups were approximately equal in size, about 300 days each. Day-of-week differences in ozone were evaluated for each group. Days with low OFP are least relevant to the issues surrounding the ozone WE effect, so emphasis is placed here on differences between the medium and high OFP groups.

For the medium OFP group, the ozone WE effect was approximately 13 to 15%. This corresponds well with the WE effect noted for high sites on all days from May through October. Under high OFP conditions, however, the ozone WE effect was approximately 7 to 8%. That is, when the meteorological conditions are most relevant to attainment goals and human health protection, ozone concentrations on weekdays and weekends differ less than usual.

This result might have been expected based on the conditions that characterize high ozone-forming potential. Studies throughout the world show that low dispersion and intense sunlight are required to reach relatively high ozone concentrations. Low dispersion is characterized by light winds combined with temperature inversions at low altitudes that trap pollutants near the surface. Intense sunlight drives the photochemical reactions that produce ozone in the lower troposphere. When intense sunlight combines with low dispersion, high surface temperatures are the typical result. High temperatures increase the rates of ozone-forming reactions that do not involve photolysis and can increase the emission rates of some ozone precursors.

Furthermore, the highest ozone concentrations measured at the surface often occur when two or more days in succession have low dispersion and intense sunlight. Under these conditions, pollutants tend to accumulate in the air and carry over from one day to the next. Days with these characteristics are likely to have an abundant supply of ozone from carryover. Some researchers conclude that carryover can contribute significantly to ozone measured at the surface the following day.[13,14] If ozone from the day before routinely contributes to surface ozone measurements, the ozone from carryover should increase during episodes with high ozone-forming potential.

Carryover of ozone along with ozone produced by carryover of precursors may routinely exceed the scavenging potential of the relatively scarce NO emissions on WE. When large amounts of ozone and other materials accumulate under episode conditions, could they cause weekdays to behave more like weekends thereby decreasing the WE effect for those days with high OFP? That would be the case if the available ozone starts to exceed the scavenging potential of the higher NO emissions on weekdays.

Since carryover may contribute a high proportion of the ozone measured at the surface on weekends and on weekdays during high-ozone episodes, what are the photochemical conditions under which these ozone molecules are generated? This question is the focus of the next section.

Ozone Carryover Aloft and the Ozone Weekend Effect

Surface ozone data show clearly that little ozone can persist at the surface from one day to the next at almost all locations in California's urbanized areas. Simultaneously, however, high concentrations of ozone and ozone precursors can carry over above the surface. Ozone-generating processes that contribute to surface ozone measurements occur routinely in the atmosphere between 100 and 1500 meters above ground level (“aloft”). These processes can generate large amounts of ozone during daylight hours. Hour by hour, some of the ozone generated aloft is carried down to the surface by convection or by turbulent mixing where it is measured on the same day. At the end of the day, a large reservoir of ozone and ozone precursors is often sequestered aloft overnight to contribute to ozone measurements and to fresh ozone generation the following day.

The physical processes that govern vertical mixing by convection and turbulence are well known. As surface temperatures increase following daybreak, convection takes place. The warm surface air rises (cooling as it does so) until it meets air of a similar temperature. The layer of mixed air at the surface thereby deepens and emissions from the surface are carried upward to mix with the air aloft.

The processes that form an overnight reservoir of pollutants aloft may not be familiar to some. Surface temperatures and mixing depths typically reach their maximum and begin to cool sometime in the middle to late afternoon. When this happens, vertical convection breaks down, the air stops mixing, and a large reservoir of ozone and precursors becomes stranded in the air aloft. The surface continues to cool overnight, as heat escapes in the form of infra red radiation. The surface cooling causes a surface-based inversion to form, and this further isolates the reservoir aloft from emissions at the surface. Ozone near the surface is almost entirely scavenged by NO emissions or destroyed by contact with materials, leading to the low overnight ozone measurements at the surface. The levels of ozone and ozone precursors aloft, however, can remain very high over night.

Table 7 presents information on ozone reservoirs aloft at 8 a.m. on intensive sampling days during SCOS97. Ozonesondes were released at five locations in the SoCAB on these days.

Under what conditions is ozone generated aloft? Data characterizing conditions at the surface are plentiful, but data characterizing conditions aloft are scarce. Measurements of conditions aloft are usually limited to intensive sampling days during special studies of regional ozone. These studies may use aircraft, balloons, remote-sensing instruments or other techniques to measure conditions aloft. The main purpose of these intensive studies is to characterize the conditions throughout a modeling domain to support simulations of the ozone-forming system. Therefore, the data are limited in their ability to illuminate the reasons for the ozone WE effect.

Despite their limitations, the available measurements aloft reveal that large amounts of ozone are generated and persist in the air aloft. Furthermore, concentrations of NO_x (or total reactive nitrogen, NO_y) decrease more quickly with altitude than do concentrations of total VOC, especially when VOC reaction products are properly included. As a result, compared to surface conditions, NO_x concentrations aloft are usually much lower, NO is virtually absent, VOC/NO_x ratios are substantially higher, and VOC reaction products are more prevalent making the VOC mix more active in producing ozone. For these reasons among others, the ozone-forming system aloft appears to be quite different from that indicated by surface data. In many ways, the photochemical conditions aloft appear to be similar to conditions rather far downwind. That is, the ozone-forming system aloft tends to look more NO_x-limited than the measurements at the surface would indicate.

Most research on the ozone WE effect has relied perforce on analyses of data collected by surface-based monitors and assumes these data represent the conditions under which ozone aloft is formed. This assumption, however, is not warranted and it may derail one's train of thought. In particular, high ozone at the <u>surface</u> on Sunday <u>does not imply</u> high carryover of ozone and ozone precursors <u>aloft</u> from Sunday to Monday. Similarly, relatively low ozone at the <u>surface</u> on Friday/Saturday <u>does not imply</u> relatively low carryover of ozone and ozone precursors <u>aloft</u> from these days to Sunday. Instead, there is every reason to assume that the loading of the atmosphere aloft at the end of the day reflects the emissions rates for that day (plus a portion from preceding days). Despite the surface data that show the greatest surface ozone on Sundays, the following analysis indicates that there is likely to be greater carryover aloft from Friday to Saturday and from Saturday to Sunday than from Sunday to Monday.

Empirical data that demonstrate or refute the preceding assertion are not now available. Nevertheless, the results in Table 8 provide some support. Table 8 reflects an analysis of ozone daily maxima from May through October in the SoCAB. For each monitoring location and each year, we determined the day of the week with the lowest ozone and tabulated the number of site-years for which the lowest ozone fell on each day of the week.

Table 8 reports separate results for coastal and inland sites for 1981 through 2000 and for four periods of five years each within that span. Through 1990, average ozone value was lowest on Monday more often than on any of the other weekdays. Since 1990, Monday has been comparable to the other weekdays in terms of the frequency with which it records the lowest average value. The significance of these results is that Monday should be expected to have the highest ozone among weekdays.

Measured ozone is highest on Sundays throughout the SoCAB, so an extra measure of ozone should carry over to Mondays and cause them to record higher ozone compared to the other weekdays. Because Monday ozone has been comparable to or lower than ozone on the other weekdays, one must question whether ozone carryover is proportional to the ozone measured at the surface. If surface data do not represent the potential for ozone carryover, perhaps the surface data do not represent the amount of ozone that forms and persists aloft.

The supposition that lower NO_x on weekends leads to greater ozone production does not seem to explain a depression of ozone on Mondays. Figure 4 indicates that heavy-duty truck traffic on weekdays may be lower on Mondays than on the other weekdays. At least until the early afternoon. In addition, Figure 5 shows that surface concentrations of NO_x may be lower throughout the day on Mondays compared to the other weekdays. If NO_x is lower on Mondays than on the other weekdays, many might expect ozone on Monday to be highest among weekdays. Instead, ozone on Mondays has been comparable to or lower than ozone on the other weekdays. Perhaps, the assumption that conditions aloft are like those measured at the surface is incorrect.

If carryover aloft plays a major role in the ozone WE effect, some vital questions must be answered. Would regulatory NO_x reductions decrease the amount of ozone generated or carried over aloft? Since weekends may well be affected more than weekdays by ozone from aloft (less ozone quenching on weekends), would WE ozone respond favorably to regulatory NO_x reductions? Since WE days increasingly determine the attainment status of California's major air basins, are regulatory NO_x reductions needed to bring these areas into attainment expeditiously?

Field studies, smog chamber experiments, and modeling exercises could all help answer these questions. Specially designed field studies could gather the data needed to describe the conditions aloft and how these change from weekdays to weekends. Field studies might even reveal the degree to which ozone generated aloft or carried over from the previous day contributes to surface ozone measurements. Smog chamber studies could start with the conditions aloft and explore the system's response to emission reductions. Alternatively, state-of-the-art simulation models could be used to investigate these relationships.

Unfortunately, all three approaches face significant hurdles. Field studies are complex, expensive, and typically require years to design, fund, and execute. Smog chamber studies may be unable to work effectively with the low concentrations of NO_x that characterize the ozone-forming system aloft. Simulation models, in their turn, have limitations discussed in the next section.

Simulation Models and the Ozone Weekend Effect

Photochemical simulation models are computer programs composed of procedures that simulate real-world processes. In particular, they simulate the chemistry and movement of ozone, ozone precursors, and their many reaction products. The algorithms and data structures that make up the procedures are, of necessity, simplified representations of real-world chemistry and meteorology. The simulation procedures require information on emissions and meteorology to be supplied from external sources (databases).

Today's state-of-the-art models are quite sophisticated. Nevertheless, the procedures and databases on which they rely cannot capture every detail of the real world, or models would cease to improve year after year. Computer speed and memory are limited, so the algorithms and data structures used in models cannot represent every detail of real-world chemistry and meteorology. Models must rely on assumptions that simplify the required calculations. Models become more detailed and simplifying assumptions become less restrictive as computers become more powerful. In addition, laboratory experiments and field studies are the basis for human knowledge concerning the chemistry and movement of pollutants. Such studies also have limitations, and the knowledge gained from them will always be incomplete.

The best available science is used to formulate public policy and today's models provide very helpful information regarding many issues. With respect to the question of the cause(s) of the ozone WE effect, however, today's models are incomplete in critical areas. They are not yet able to provide confident assessments of the separate contributions of multiple potential causes. Some significant limitations are discussed further in the following paragraphs.

The chemical mechanisms available today do not yet include some potentially important reactions. "Renoxification" of nitric acid, and some other recently discovered reactions, might cause NO_x emissions to produce more ozone than presently indicated using today's chemical mechanisms. In addition, the experiments on which chemical mechanisms are based have rarely included studies in which the initial conditions resembled the daytime or nighttime conditions aloft (100 to 1500 m above the ground). Compared to conditions near the surface, the air aloft is characterized by significantly higher VOC/NO_x ratios, low NO_x levels, and extremely low NO levels. No published chemical mechanism has been fully validated under such conditions. Until recently, few (if any) smog chambers could handle the required experiments. Facilities suitable for low-NO_x experiments have only been available in recent years, and more are under construction. As chemical mechanisms are updated to reflect new information, it is quite possible that ozone formation aloft and/or carryover aloft will become increasingly important when modeling the ozone WE effect.

Processes that move and/or dilute pollutants include turbulent mixing (mechanical and convective), advection into and out of a region (including recirculation), and deposition that moves pollutants out of the atmospheric system. The simulated movements of pollutants are largely determined by "wind fields" generated during a modeling exercise. Vertical mixing may be systematically understated by some systems that generate wind fields. Vertical mixing may also be suppressed by interactions between wind fields and the vertical layers used in models to represent the atmosphere at various distances above the ground. In such cases, the model may not capture the real-world impact of ozone that forms aloft and later mixes down to the ground.

Most models include vertical layers that increase in thickness as the height above the ground increases. This feature is often dictated by the limited power of computers, which may not be able to process tens or hundreds of equally thin layers in a reasonable amount of time. A side effect of this practice, however, may be to dampen the impact of vertical mixing on ozone measured at the surface of the ground. When pollutants, such as ozone precursors, move upward from thinner cells into a thicker cells, their masses are artificially diluted as they are dispersed in larger volumes. This has the desirable effect of preserving mass as materials move between cells in a model, but the previously diluted materials may produce too little ozone aloft. Therefore, when artificially small amounts of materials mix downward from thicker cells to thinner cells, their impact on pollutant levels at the surface may be artificially small.

Some WE emission inventories have already been developed, and these continue to improve. Nevertheless, uncertainties regarding WE inventories remain at least as great as the uncertainties regarding WD inventories. Similarly, WE episodes from intensive field studies are still rather limited. Although WE episodes were obtained during the 1997 Southern California Ozone Study, simulations are still being carried out on the most promising of these episodes. For these simulations, satisfactory model performance, particularly for estimating pollutant levels aloft, is not yet established. Because pollutant measurements aloft are very limited, model performance in this area may not be possible to assess adequately.

For the preceding reasons, among others, we consider the models and the input data available today to have significant limitations relative to determining the specific cause(s) of the ozone WE effect. Even so, the models do provide useful information and future improvements in both the models and model inputs may help to better understand the ozone WE effect.

CONCLUSIONS

The following questions were posed before the presentation and discussion of analyses of the ozone WE effect.

1. When ozone molecules are detected at a surface monitor,
   - Where were they generated?
   - When were they generated?
   - Under what conditions were they generated?
2. How do the answers to these questions differ on weekdays versus weekends?
3. What implications do these differences have regarding regulatory (steady) reductions of VOC and NO_x rather than WE (intermittent) reductions of VOC and NO_x.

Conclusive answers to all of these questions are not yet available because they depend on tools and/or data that are not sufficiently complete or reliable. Nevertheless, the analyses in this paper provide a coherent set of plausible, perhaps probable, answers.

Ozone molecules detected by surface monitors are generated in various locations under various conditions. People usually consider two locations – near field and downwind – that are oriented horizontally when pondering the impact of the ozone precursors, VOC and NO_x. However, two other key locations – "near the surface" and "aloft" – are oriented vertically, rather than horizontally. Conditions aloft, between 100 and 1500 meters above ground level, have much in common with conditions "downwind." In particular, these conditions include low NO_x (commonly < 20 ppb) and high VOC/NO_x ratios, with NO virtually absent. Ozone formation under these conditions is much more NO_x-limited than surface measurements would indicate. Therefore, the surface data cannot be assumed to represent conditions aloft.

Large reservoirs of ozone and ozone precursors can accumulate and carryover from one day to the next. This phenomenon is especially important when dispersion is low and solar intensity is high, the very conditions that lead to high ozone episodes. During such episodes, a large proportion of ozone measured at the surface is likely to have been formed aloft under conditions that appear to be rather NO_x-limited. That is, reduced NO_x emissions might be expected to reduce the amount of ozone formed aloft. This in turn would reduce the contribution of ozone aloft to ozone measured at the surface.

Four different types of data indicate that a significant portion (perhaps, the major share) of the ozone WE effect may be due to differences in ozone quenching very near ground level. High NO emissions on weekdays quench (i.e., destroy) large amounts of ozone near the ground. When NO emissions decrease on weekends, the surface-level ozone quenching decreases and WE ozone measurements at the surface increase relative to the WD levels. This surface phenomenon cannot be assumed to apply to ozone formation aloft, however, where reduced WE NO_x emissions may reduce rather than increase the amount of ozone present.

Long-term ozone trends indicate that these observations and conjectures may explain a great deal of the ozone WE effect. Thirty years in which the rate of VOC reductions has exceeded the rate of NO_x reductions have seen greater ozone reductions on weekdays than on weekends. Weekends have been left behind to such an extent that WE days now play the largest role in determining the attainment status of major air basins in California. In the air basins that have been carefully studied, the ozone WE effect is relatively small but may be increasing at the downwind receptor sites that determine the attainment status of each air basin.

There is every reason to anticipate that a continuing emphasis on VOC emission reductions will further widen the gap between weekdays and weekends as weekends improve at a slower rate. The evidence presented here reasonably supports the conclusion that NO_x-reductions are important and could accelerate the rate of improvement for ozone on all days, including weekends.

DISCLAIMER

The statements and conclusions in this paper are those of the author and not necessarily those of the California Air Resources Board. The mention of commercial products, their source, or their use in connection with material reported herein is not to be construed as actual or implied endorsement of such products.

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Table 1. Annual average VOC and NO_x emissions inventory* (tons per day) for 1980 and 2000 in four regions of California.

Region	Pollutant	Year 1980	Year 2000	Percent Decrease
South Coast Air Basin	VOC	2218	1029	54 %
	NO_x	1743	1131	35 %
Bay Area Air Basin	VOC	1333	552	59 %
	NO_x	966	636	34 %
Sacramento Metro AQMD	VOC	189	97	49 %
	NO_x	133	109	18 %
San Joaquin Valley Air Basin	VOC	1032	477	54 %
	NO_x	775	577	26 %

* From ARB database supporting emissions projections for the 2002 Almanac of Emissions and Air Quality, California Air Resources Board, Planning and Technical Support Division, Sacramento, CA.

Table 2. Rates of progress reducing ozone concentrations in the South Coast Air Basin from 1980 – 1998 based on the average of the 2nd through 11th highest values for daily maximum ozone by day of week.

Sub-Region	Sites Used	Weekdays*	Weekends*
All sites	17	Down 46 %	Down 33 %
Southwest L.A. County	4	Down 46 %	Down 34 %
San Gabriel Valley	3	Down 55 %	Down 36 %
San Fernando Valley	2	Down 49 %	Down 43 %
Orange County	3	Down 43 %	Down 26 %
San Bernardino/Riverside	5	Down 42 %	Down 31 %

* Difference between the 1996/98 and 1980/82 values expressed as percent change with respect to the 1980/82 baseline. Weekday values represent Monday through Friday.

Table 3. Percent change in ozone from Friday to Sunday at Urbanized versus Suburban/Rural sites in four regions of California (data for May-October 1998-2000).

	Average Change in Ozone – Friday to Sunday			
	Urbanized Sites		Suburban/Rural Sites	
Region	Sites	Average	Sites	Average
South Coast Air Basin	12	29.3%	7	19.0%
San Francisco Bay Area Air Basin	15	26.1%	7	20.7%
Sacramento Metropolitan Area	3	10.5%	6	4.8%
San Joaquin Valley Air Basin	13	8.6%	6	4.4%

Table 4. AIRS ID numbers for sites classified as Urban or as Suburban/Rural in four regions of California.

Region	Urban		Suburban/Rural	
South Coast Air Basin	060590001	060370002	060710005	060712002
	060371002	060370016	060659001	060714003
	060595001	060371103	060658001	060719004
	060371301	060372005	060376002	
	060371601	060371701		
	060371201	060711004		
San Francisco Bay Area Air Basin	060130002	060011001	060131002	060950002
	060012001	060851001	060850002	060010007
	060851002	060010005	060010003	060550003
	060133001	060811001	060852006	
	060850004	060852005		
	060010006	060131003		
	060410001	060970003		
	060950004			
Sacramento Metropolitan Area	060670002	060670006	060610002	060170020
	060670010		060670012	060170010
			060613001	060610006
			060675003	
San Joaquin Valley Air Basin	060290014	060290010	060295001	060290007
	060195001	060190008	060311004	060290008
	060190007	060190242	060194001	060773003
	060990005	060990010		
	060290232	060190010		
	060770009	060771002		
	061072002			

Table 5. Differences on weekdays and weekends between surface ozone and ozone at 100 m above ground level expressed as percent of ozone at 100 m; results based on ozonesonde data at 2 p.m. during the 1997 South Coast Ozone Study.

	Weekdays		Weekends	
Location	**Average**	**Maximum**	**Average**	**Maximum**
Anaheim	22% (9)*	78%	13% (4)	17%
Los Angeles	9% (7)	16%	8% (4)	10%
Northridge	17% (8)	34%	13% (4)	20%
Pomona	23% (9)	35%	17% (4)	23%
Riverside	20% (6)	62%	9% (4)	11%

* The number of 2 p.m. ozonesondes available is given in parentheses next to the average.

Table 6. Comparison of total oxidant (O_3 plus NO_2) at locations in the South Coast Air Basin. Values are means of daily maxima from 10 a.m. to 6 p.m. for May – October from 1998 through 2000.

Location	Total Oxidant (ppb) WD	Total Oxidant (ppb) WE	Difference (% of WD)
Hawthorne	69	71	2.5%
Pasadena	98	103	4.4%
Pico Rivera	97	102	6.1%
San Bernardino	107	116	8.5%
La Habra	85	90	6.9%
Fontana	105	114	8.6%
Reseda	78	79	0.9%
N. Long Beach	76	79	3.6%
Azusa	110	118	7.2%
Upland	109	119	8.7%
Burbank	104	105	0.6%
West L.A	67	69	2.9%
Lynwood	75	78	4.3%
Riverside	94	100	7.2%
Anaheim	78	83	6.7%
Glendora	109	117	7.6%
Santa Clarita	92	94	1.6%
Pomona	106	114	8.1%
Los Angeles - N. Main	91	94	3.0%
Costa Mesa	63	65	3.7%
Lake Elsinore	88	91	3.5%
Banning	95	95	-0.8%
Basin Average			**4.8%**

Table 7. Carryover of ozone* (ppb) formed the previous day and sequestered in a layer of polluted air above the ground surface. Pollutants within 1500 m of ground level are likely to contribute to pollutant measurements at the surface the next day.

Location	Number of Days	Ozone 200 to 1000 m** Avg.	Min	Max	Ozone 200 to 1500 m** Avg.	Min	Max
Anaheim	13	45	24	61	48	30	64
Central Los Angeles	11	44	25	62	48	32	65
CSU Northridge	13	51	35	71	52	39	72
Pomona	13	50	21	68	51	27	62
Riverside	13	47	23	67	47	28	64

* Data from 8 a.m. ozonesondes during the 1997 Southern California Ozone Study
** Elevation above ground level

Table 8. Average max ozone and percent of site-years* with which different days of the week recorded the lowest mean of daily maximum ozone measurements during the May to October ozone season in California's South Coast Air Basin.

			Day of Week						
Area	**Period**	**Summary for Site/Years**	**Sun**	**Mon**	**Tue**	**Wed**	**Thu**	**Fri**	**Sat**
Coastal: 15 sites	1981 - 1985	Average (ppm)	0.113	0.102	0.106	0.109	0.109	0.111	0.117
		% lowest	1%	51%	8%	7%	25%	6%	1%
	1986 - 1990	Average (ppm)	0.103	0.092	0.096	0.095	0.094	0.095	0.102
		% lowest	3%	33%	13%	19%	23%	9%	0%
	1991 - 1995	Average (ppm)	0.095	0.076	0.076	0.077	0.078	0.080	0.092
		% lowest	0%	43%	11%	13%	15%	19%	0%
	1996 - 2000	Average (ppm)	0.076	0.059	0.057	0.058	0.056	0.060	0.072
		% lowest	0%	13%	24%	30%	18%	15%	0%
Inland: 5 sites	1981 - 1985	Average (ppm)	0.130	0.130	0.135	0.140	0.138	0.141	0.139
		% lowest	33%	38%	0%	4%	17%	4%	4%
	1986 - 1990	Average (ppm)	0.123	0.124	0.130	0.130	0.132	0.131	0.128
		% lowest	36%	24%	8%	12%	12%	0%	8%
	1991 - 1995	Average (ppm)	0.119	0.106	0.106	0.107	0.108	0.111	0.124
		% lowest	4%	48%	8%	12%	16%	12%	0%
	1996 - 2000	Average (ppm)	0.096	0.080	0.079	0.081	0.079	0.079	0.093
		% lowest	0%	20%	20%	28%	16%	16%	0%

* The maximum number of "site-years" possible is the product of the number of sites and the number of years in the period. For each site in each year, the day of week with the lowest average for daily max-hour ozone was identified and tallied. The frequency for each day of the week is expressed as a percent of the total number of site-years available.

Figure 1. Ozone trends from 1980 to 1998 on weekdays (dashed lines) and weekends (solid lines) at selected sites in the South Coast Air Basin. Values are based on day-of-week ozone levels using the 2nd through the 11th highest daily maxima per year for each day of the week. For each day of the week, about 33% of the days during the ozone season are included in the trends.

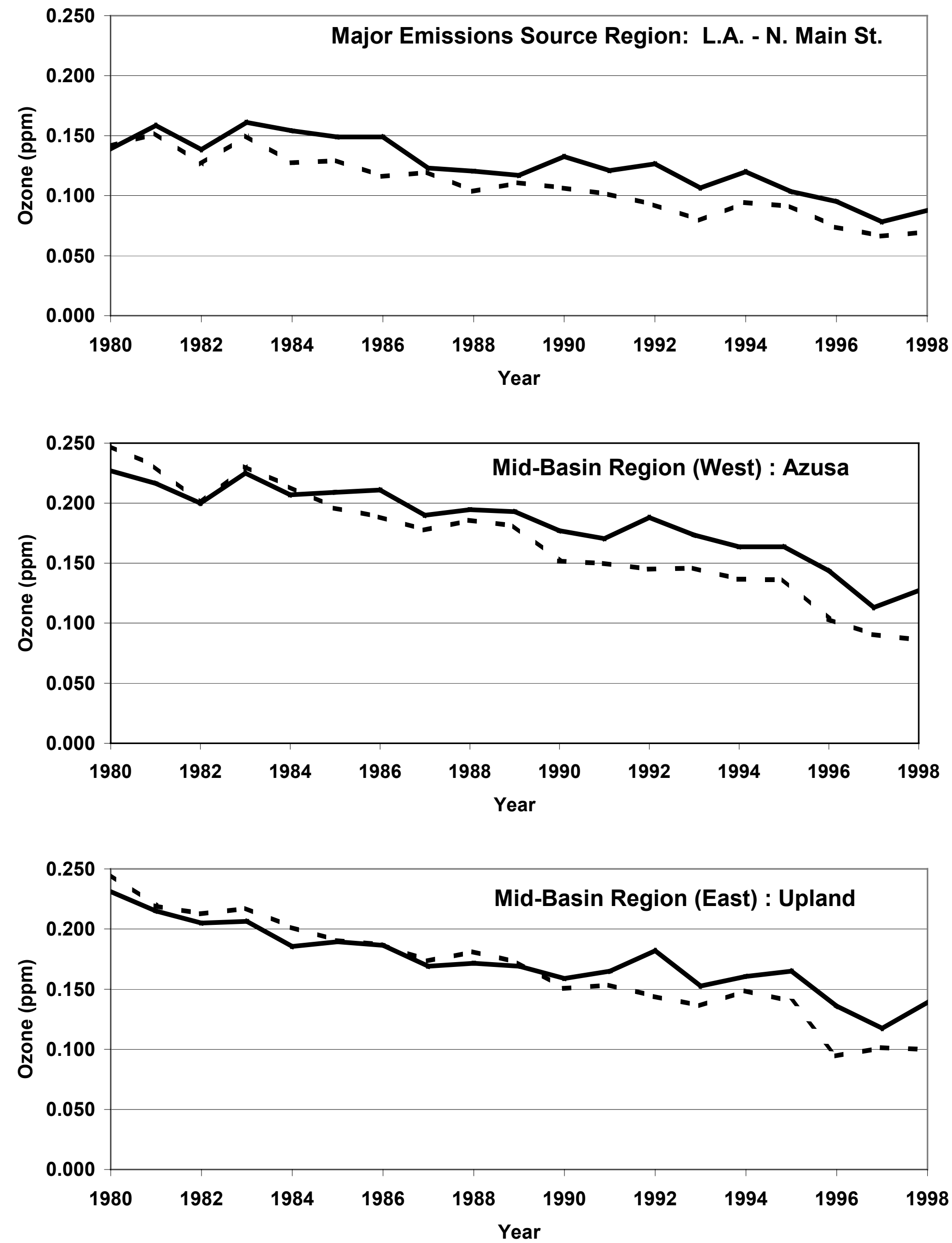
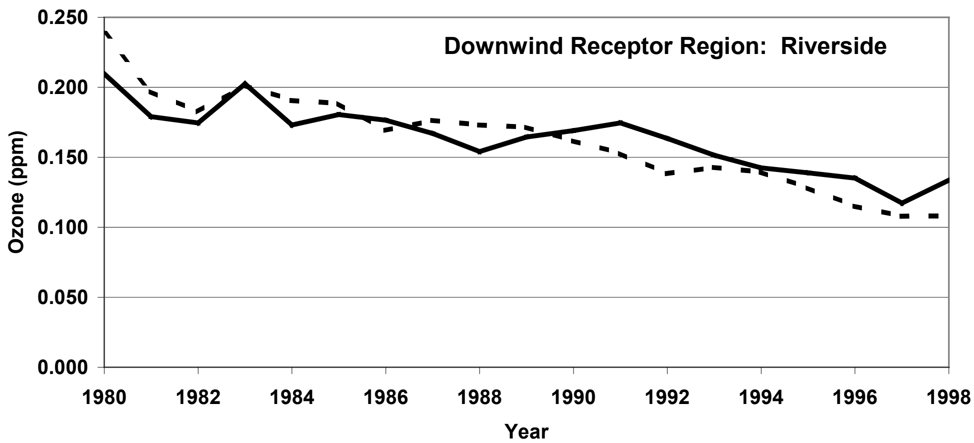


Figure 1 (continued). Ozone trends from 1980 to 1998 on weekdays (dashed lines) and weekends (solid lines) at selected sites in the South Coast Air Basin. Values are based on day-of-week ozone levels using the 2nd through the 11th highest daily maxima per year for each day of the week. For each day of the week, about 33% of the days during the ozone season are included in the trends.



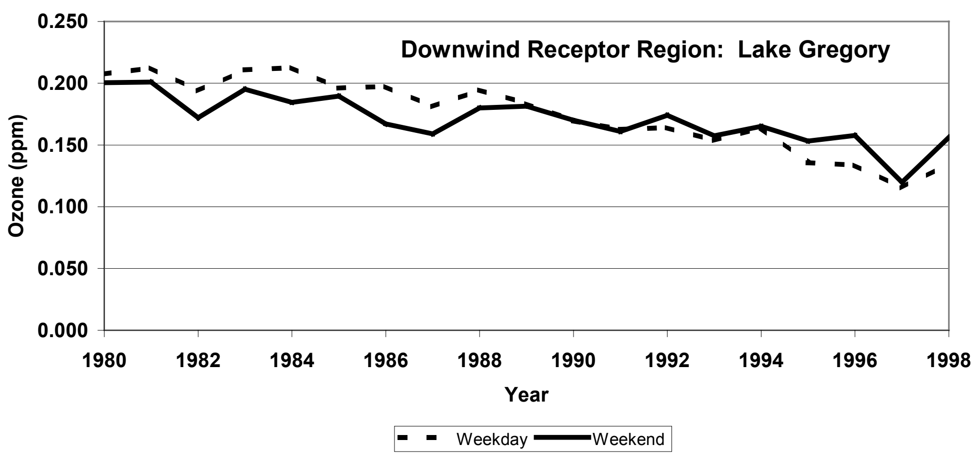
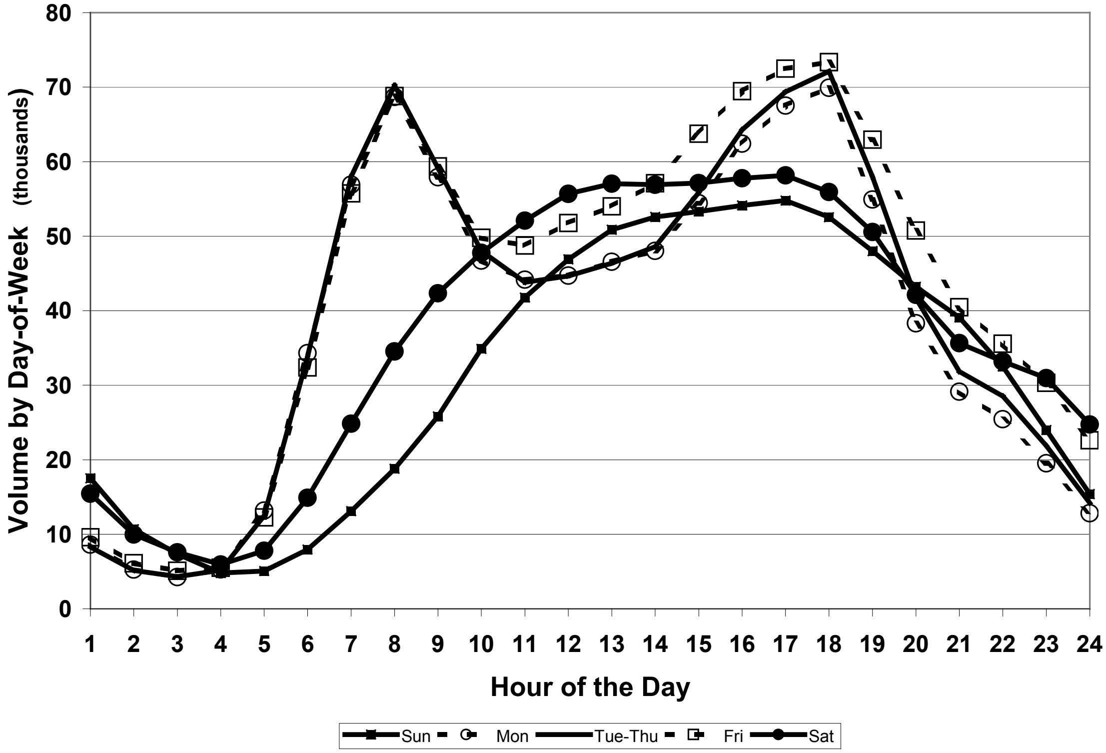


Figure 2. Freeway activity of light-duty vehicles by day-of-week – composite of 2000 data from 11 Weigh-in-Motion stations in California's South Coast Air Basin.



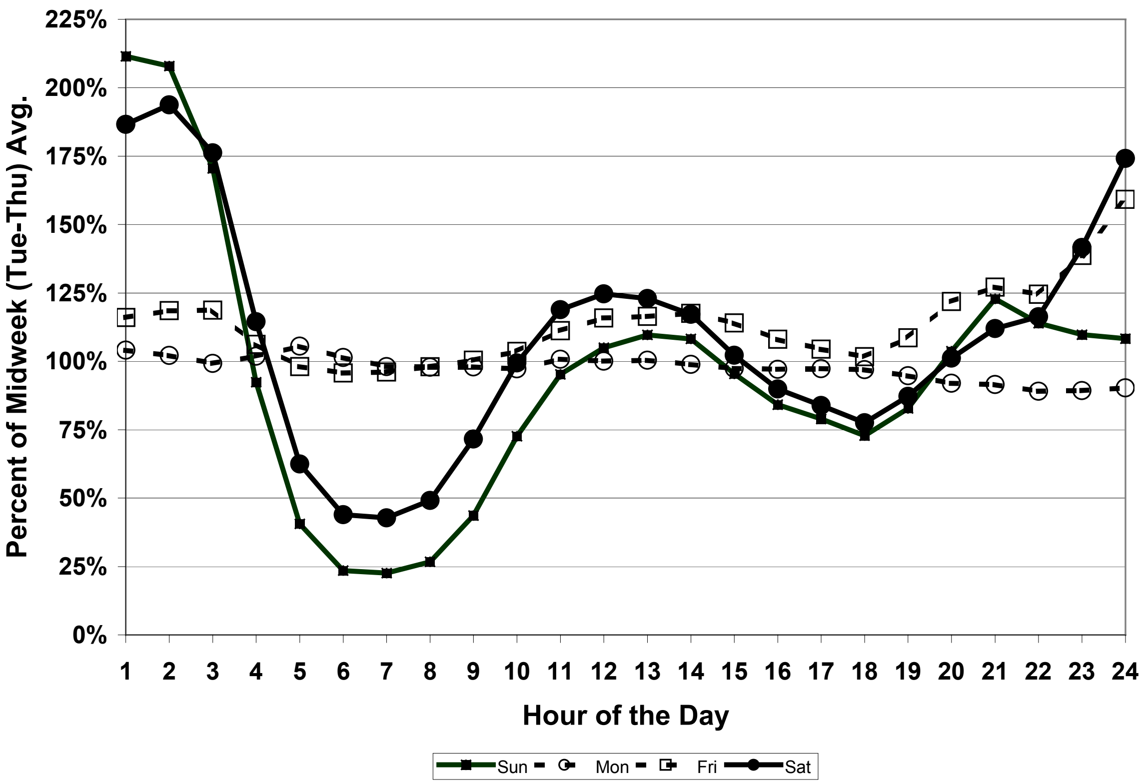
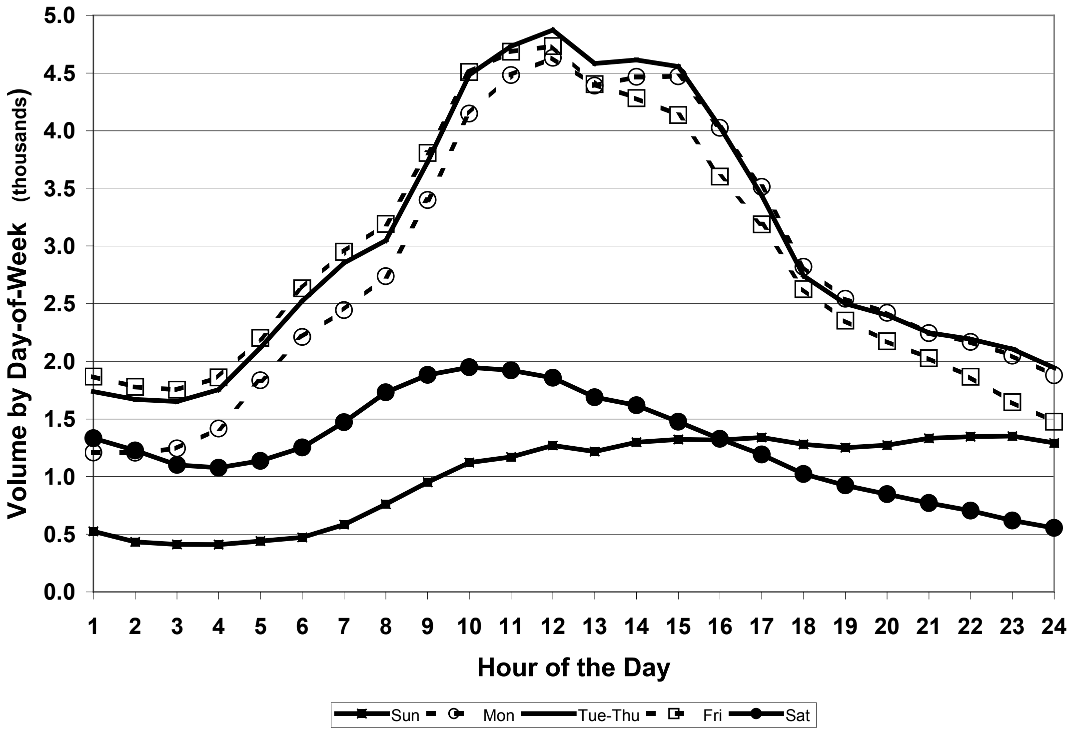


Figure 3. Freeway activity of heavy-duty vehicles by day-of-week – composite of 2000 data from 11 Weigh-in-Motion stations in California's South Coast Air Basin.



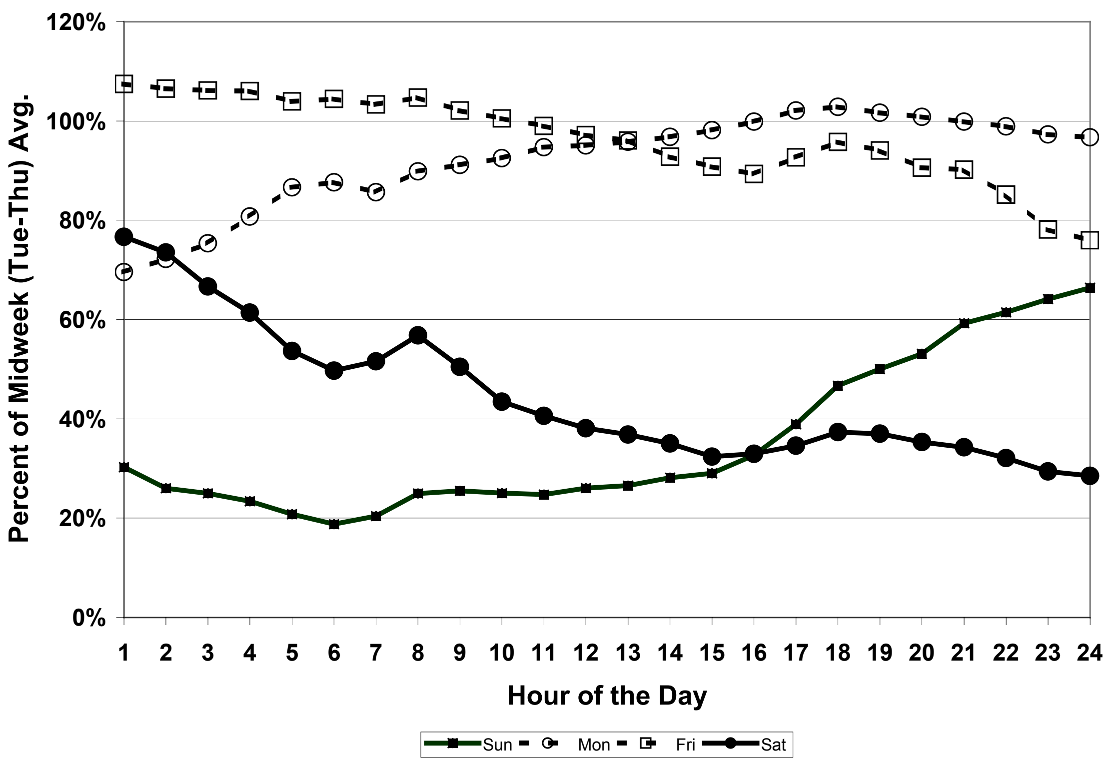
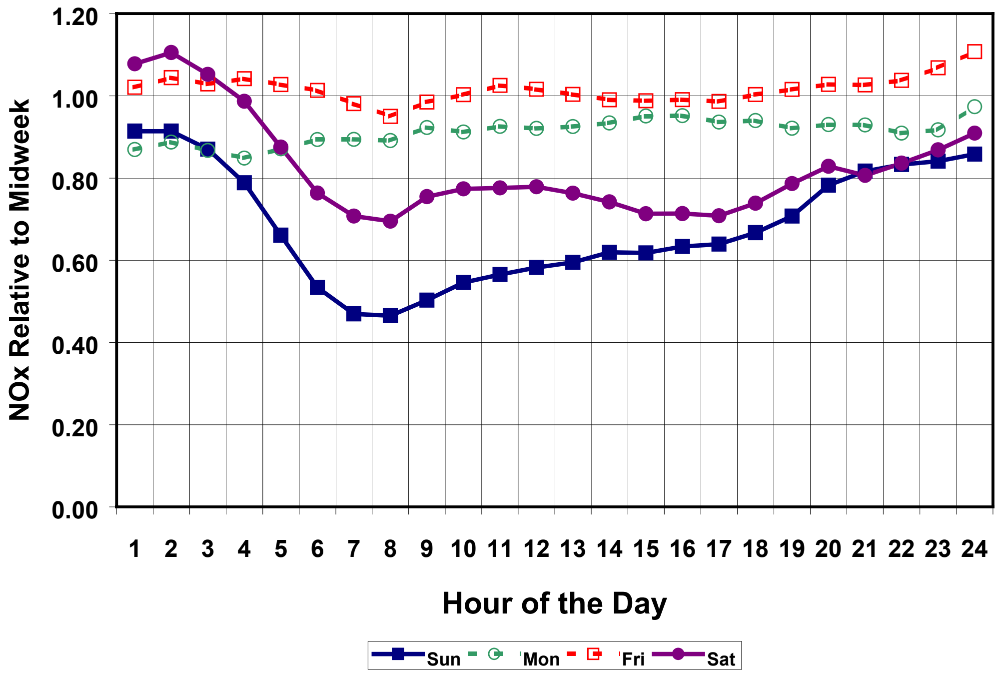


Figure 4. Hourly values of NO_x by day-of-week expressed as a percent of midweek (Tue-Thu) value; composite of 11 sub-regions of the South Coast Air Basin



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