

OXIDANT MEASUREMENTS FROM THE SOUTH COAST AIR BASIN:
CONSIDERATIONS FOR CONTROL STRATEGIES

by

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At the November 14, 1974 meeting of the ARB^{*}, a preliminary report concerning oxidant calibration procedures was presented by the Oxidant Measurement Committee. The report indicated that measurements of oxidant by the ARB were 29% high, and measurements by the LACAPCD^{*} were 4% low as compared to the ultraviolet spectrophotometric method. The ratio of measurements by the ARB to the LACAPCD were therefore: $\frac{1.29}{.96} = 1.34$.

In other studies the ratio has ranged from 1.3 to 1.7. When considering the average ratio as determined by personnel who have had considerable experience in calibration procedures, the margin is somewhat narrower. Results by the AIHL indicated a ratio of 1.4. In a joint study by the ARB and the LACAPCD, the ratio was 1.5. When an instrument of the ARB was compared directly to measurements with an LACAPCD instrument, the ratio was 1.33. At Azusa, where an ARB Dasibi was compared to a LACAPCD KI instrument over a period of six weeks, the ratio was 1.45; the ratio of the max hour concentrations was approximately 1.35.

The oxidant concentrations measured by the LACAPCD that are shown in the following figures were adjusted by a factor of 1.35 to make them equivalent to measurements by the ARB procedure. It would have been better to adjust by a factor of 1.4 because the calibration data are not accurate to three significant figures. To obtain the true oxidant concentrations,

*ARB--California Air Resources Board

*LACAPCD--Los Angeles County Air Pollution Control District

subtract 1/5 of the values shown in Figure 1 ($1/1.29 = .78$). This assumes that measurements by the ultraviolet spectrophotometer method represent true ozone concentrations.

Figure 1 is from the ARB report "Oxidant Trends in the South Coast Air Basin." It shows the average of the daily max one-hour concentrations during the July, August and September months, 1970-72. The data in this figure have not been adjusted. The isopleths show an increasing ozone concentration in a northeastwardly direction from the coast. This figure may have been the basis of the statement in a recent National Academy of Sciences report that oxidant concentration increased steadily in an eastwardly direction.

Figure 2 shows the isopleths and the concentrations after adjusting the LACAPCD data by the factor 1.35. As shown, the highest concentrations are now in the Pasadena-Azusa area. Further east of this area, the concentrations decrease. It should be noted that there were no data from the Upland-Fontana area and the isopleths are based on data from about 15 air monitoring stations.

By 1974 (Figure 3) the number of air monitoring stations in the area shown on the map had nearly doubled, so that the ozone isopleths shown have greater reliability than those on the previous figures. The area of the highest concentration extends from Upland to Fontana.

The stations at Mt. Lee and in Temple City are ARB stations. Considering the sensitivity of the KI calibration procedures, the data are in remarkably good agreement with the adjusted data at nearby LACAPCD stations.

During the three months, two oxidant analyzers were in operation at Upland, and also at Fontana and Riverside. The data again were in reason-

ably good agreement although the instruments were not the same types. The instrument types used were:

Upland	-	KI and Chemiluminescence
Fontana	-	U.V. Photometry (ARB) and Chemiluminescence
Riverside	-	KI and U.V. Photometry

The previous figures dealt with average max hour concentrations during the smog months. Figure 4 shows the concentrations and isopleths for June 21, 1973. The weather this day was atypical. Light easterly winds prevailed. As shown, instead of being in the Upland-Fontana area, the highest oxidant concentrations were in Los Angeles.

In 1974, the severest smog occurred on June 27 (Figure 5). Weather conditions were typical for smog days. The highest concentrations extended from Azusa to Fontana and the isopleths had about the same pattern as in Figure 3.

Three concentrations are shown at Upland - 55 pphm as measured by a Dasibi instrument; 57 as measured by the KI method; and 63 as measured by a chemiluminescent instrument which was approximately two miles from the site of the other instruments. It is uncertain whether or not the third stage episode criteria level was exceeded. The highest concentration, 63 pphm, measured at the San Bernardino County APCD exceeded the third stage level but the average of the three stages was below the level.

Figure 6 is from the work by Basil Dimitriades. It shows the maximum ozone concentrations on the Y axis, NO_x concentrations on the X axis, and isopleths of non-methane hydrocarbon concentrations. Depending on the ratio of hydrocarbon to NO_x , as well as absolute concentrations, a reduction in hydrocarbons may cause a marked decrease in the oxidant concentrations or

may cause no reduction at all. For example, at the point where the 3.2 and the 2.1 ppm C isopleths intersect, there would be no reduction in oxidant when the hydrocarbons are reduced from 4.7 ppm C to 3.2 ppm C. At a NO_x concentration of .6 ppm, reducing hydrocarbons from 3.2 to 2.1 produces a more than proportional reduction of oxidants.

Superimposed on the isopleths are the 6 to 9 a.m. concentrations of hydrocarbons and oxides of nitrogen that were measured in Downtown Los Angeles for five days. The wind projection data show that on these five days the air mass passed through the downtown area to Pasadena. The maximum oxidant concentrations measured at Pasadena and adjusted by the factor 1.35 are also shown. When the concentrations of hydrocarbons and oxides of nitrogen in the chamber are about the same as the 6-9 a.m. concentrations in Los Angeles, the oxidant concentrations formed in the chamber are in good agreement with the Pasadena max hour concentration.

The lower envelope shows where we will be in 1985 if the hydrocarbon and oxides of nitrogen are reduced as projected in the control program. We will still be above the oxidant standard, and we are in an area where increasing NO_x concentrations will cause a decrease of oxidant concentrations.

The ratio of hydrocarbons to NO_x for the five days ranged from 5 to 12 (ppm C to ppm NO_x). It is not unusual for the ratio to vary by a factor of 4 from day to day. The basinwide average can vary by a ratio of two. To explain this better, variation of ratios, emission data and the spatial and temporal distribution of the sources are needed.

Table I shows the emission inventory data for reactive hydrocarbons from the year 1965 to 1985. As indicated, there has been a steady decrease of hydrocarbon emissions since 1965 and the decrease is expected to continue

through 1985.

The absolute values of the hydrocarbon emission estimates are not firm. For example, the emissions from mobile sources were based on the CVS 2 cycle. In an earlier report, the emissions were based on the 7 mode cycle and were considerably different than the values shown. It is questionable whether the 7 model or the CVS 2 represent the actual emissions during the variety of driving conditions in the South Coast Air Basin.

Table II shows that oxides of nitrogen concentrations increased from 1965 thru 1972. There is a slight decrease projected thru 1985. Again, the absolute values are uncertain. A recent study by KVB indicated, for example, that the stationary source emissions are perhaps from 15 to 20% higher than those shown in the table.

As indicated earlier, the concentrations of oxidant should decrease with a decrease in ratio of hydrocarbons to oxides of nitrogen. Table III shows that with the state control strategy, the ratios have been decreasing and will continue to decrease thru 1985.

The three-year moving mean of the daily max NO_x concentrations from 1966 thru 1973 are shown in this Figure 7. As indicated, the concentrations in the Burbank-Reseda-Los Angeles area increased from the three-year period, 1965-1967 thru the three-year period, 1970-72, and have decreased slightly since. The same trend is observed in the Azusa-Pasadena-Pomona area, and qualitatively is in agreement with the emission inventory data.

The three-year moving mean of the oxidant concentrations during the smog months have steadily decreased from the coastal cities - Lennox, Long Beach and West Los Angeles (Figure 8). From the Burbank-Reseda-Los Angeles area, the concentrations decreased until the 1971-73 period and then increased.

The increase could be attributed to the decrease of the NO_x concentrations that began in about 1972.

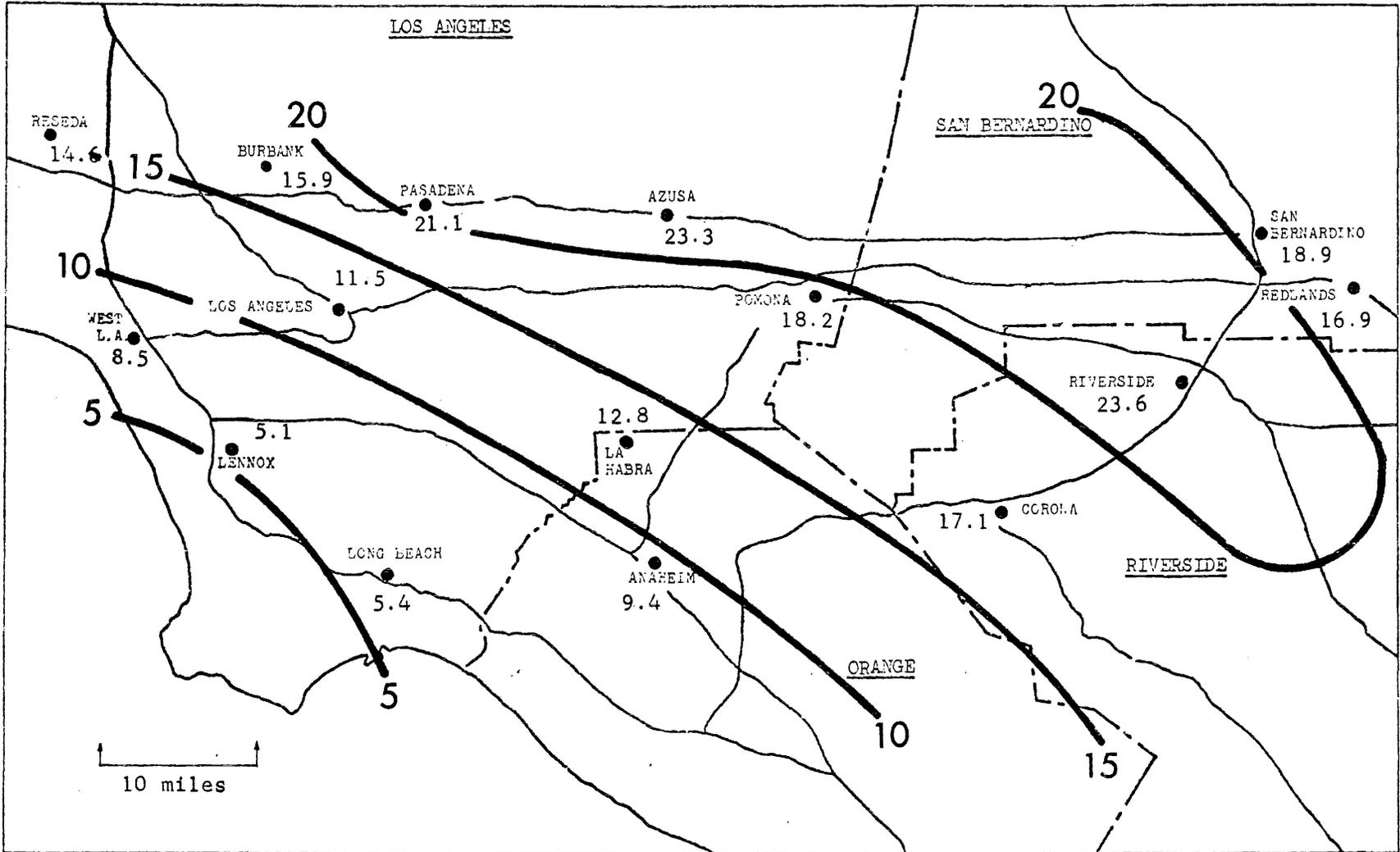
While oxidant concentrations have decreased in the coastal cities and in the major source areas, the concentrations increased at Azusa, Pasadena and Pomona until about 1970 and then decreased markedly (Figure 9). In the Riverside-San Bernardino area, concentrations appear to have steadily increased since 1965. Both the adjusted and unadjusted concentrations are indicated in the figure for the Azusa-Pasadena-Pomona area. Without the adjustment, the highest concentrations would have been in the Riverside-San Bernardino area. With the adjustment, the highest concentrations remain in the Azusa-Pasadena-Pomona area. Unfortunately we do not have continuous data from 1965 thru 74 for the Upland-Fontana area where the concentrations today are the highest in the basin. As a guess, we would predict that the trend in these areas are in between those of the surrounding areas. Based on this assumption, there has been a slight decrease in recent years.

The trends shown in Figures 8 and 9 were based on three year moving means of the daily max hour concentrations through 1972. In the ARB report "Oxidant Trends in the South Coast Air Basin," it was pointed out that concentrations in the Riverside area appeared to have peaked in 1970. With two more years of air monitoring data - 1973 and 1974, it now appears that oxidant concentrations in the far inland areas of the South Coast Air Basin are not declining. The table below shows that concentrations in 1973 and 1974 were in general substantially higher than in 1972.

AVERAGE OF DAILY MAX HOUR OXIDANT CONCENTRATIONS
July-August-September months

	<u>1972</u>	<u>1973</u>	<u>1974</u>
Azusa	24.6	25.8	27.2
Pomona	19.8	21.3	23.9
Pasadena	23.1	23.6	24.3
Upland	--	28.0	31.6
San Bernardino	14.9	20.8	26.0
Redlands	13.7	--	24.5
Riverside			
(Rubidoux)	--	19.1	23.5
Riverside			
(Magnolia)	--	20.1	20.5

The increase may be attributed to the lack of NO_x control. Still other factors such as the rapid growth in the eastern portion of the basin and the change of the relative amounts of various hydrocarbon species may be responsible. To understand why the control program for hydrocarbons has not been effective in all areas of the basin, better information on emissions and the temporal and spatial distribution of the sources and of the concentrations in air of each species of organic gas, as well as of NO_x and intermediate smog products, is necessary. Additionally, environmental chamber studies to determine the kinetics and mechanisms of the reactions are needed. Finally, research to develop three-dimensional simulation models is a must for development of control strategies that are effective for all areas of the basin.

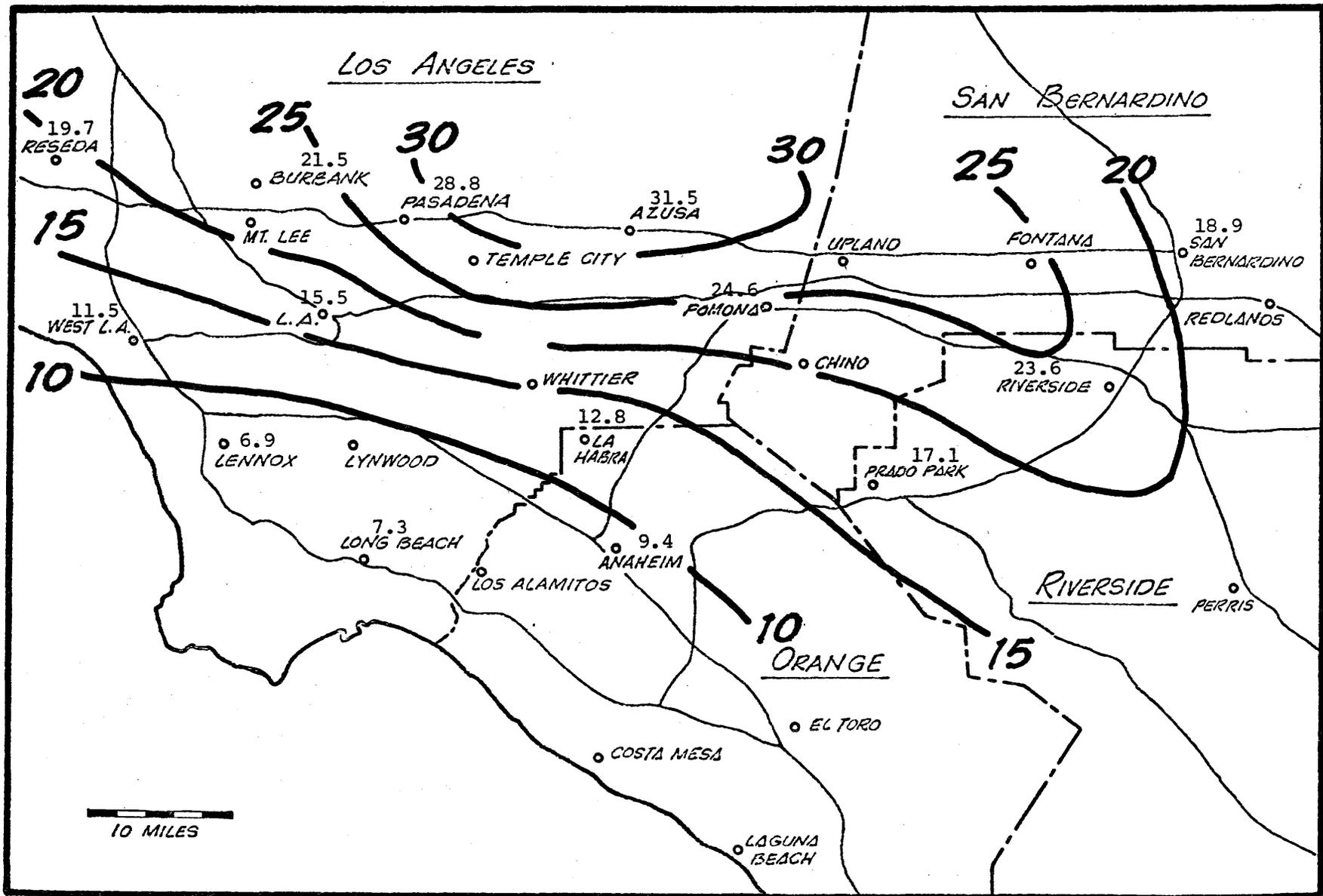


DISTRIBUTION OF OXIDANT CONCENTRATIONS

Average of Daily Maximum One-Hour Oxidant Concentrations (pphm)

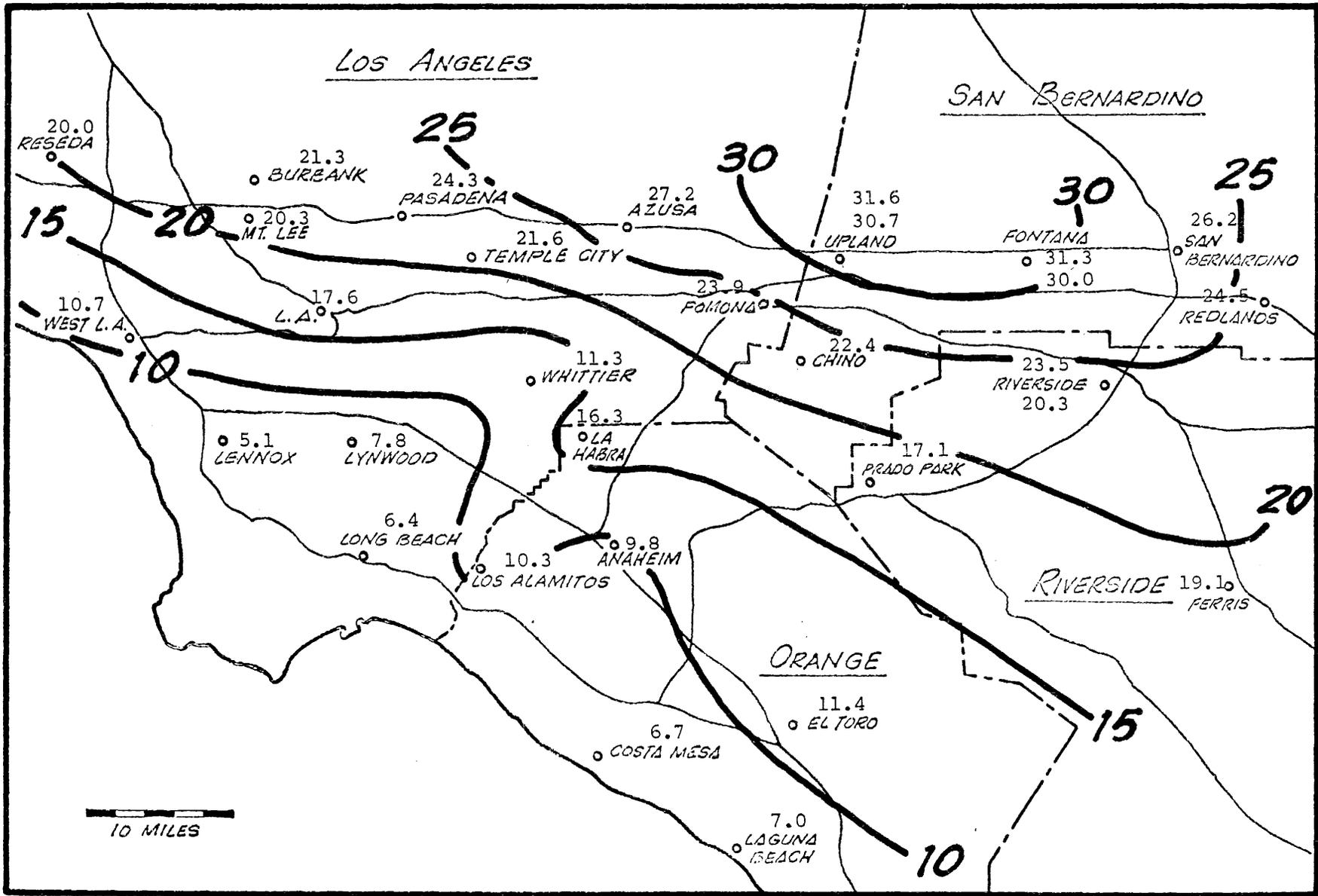
During July, August & September, 1970-1972

Figure 1



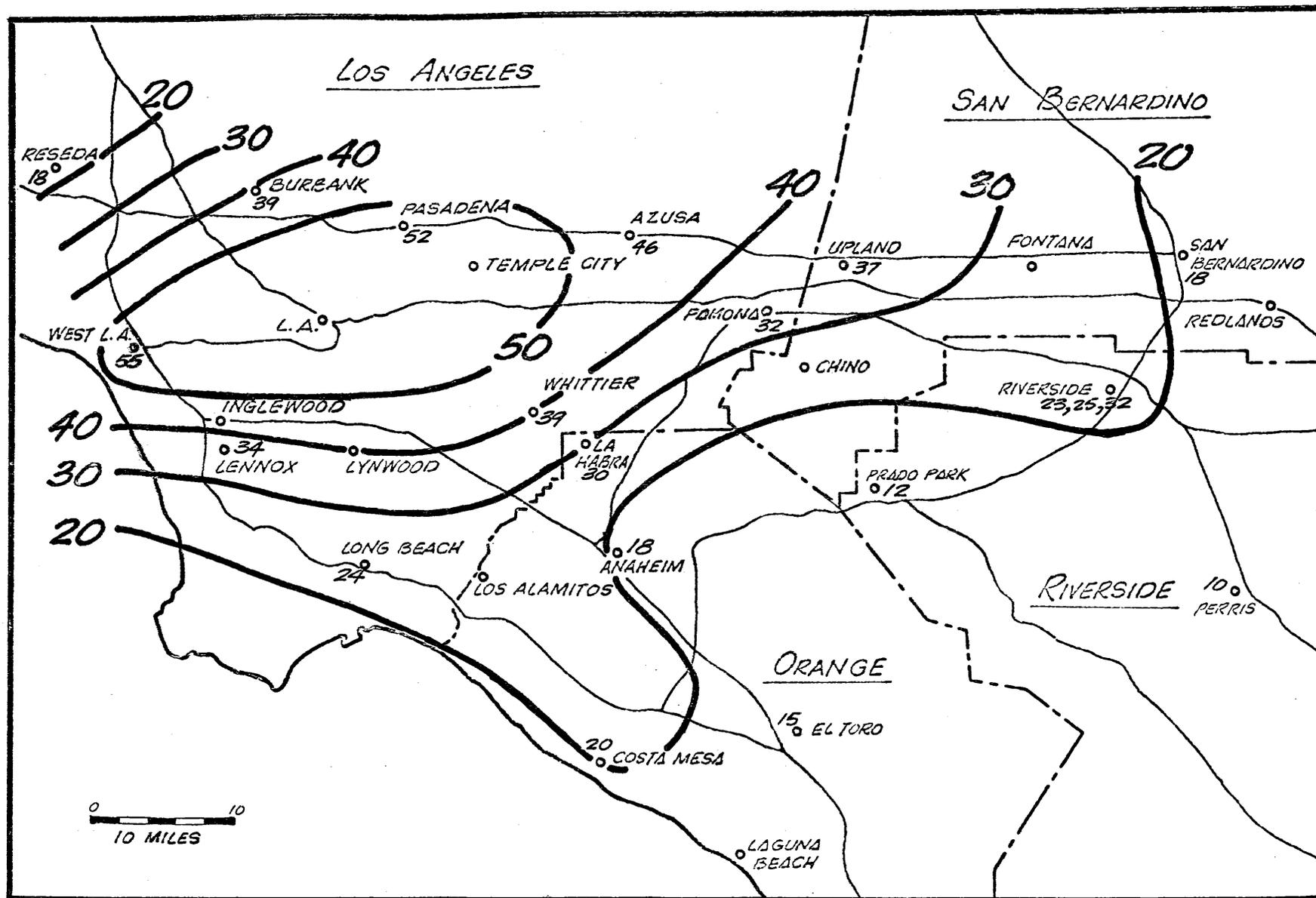
Average of Daily Max. One-Hour Oxidant Concentrations (pphm)
 During July, August, September 1970-1972 (normalized)

Figure 2



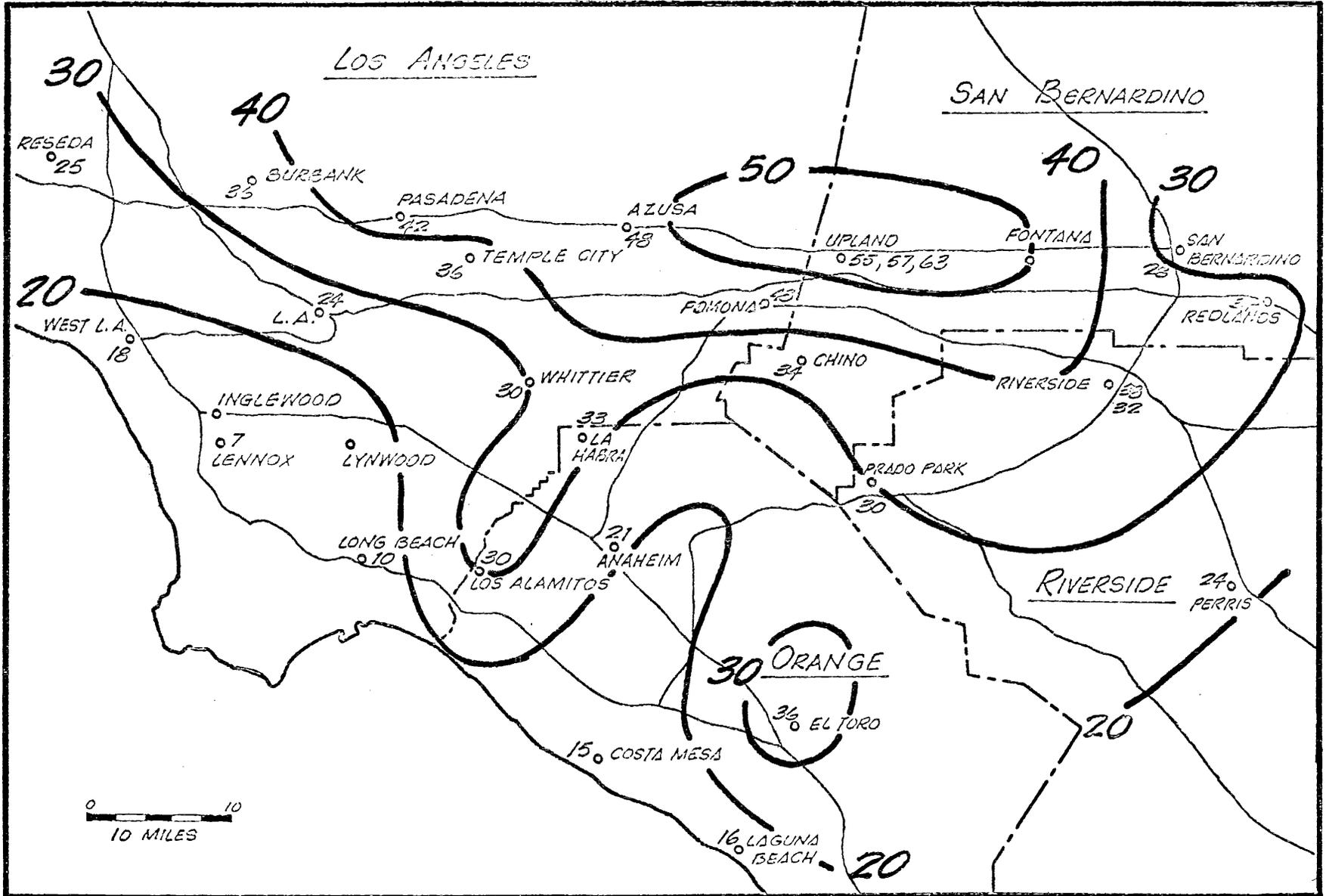
Average of Daily Max. One-Hour Oxidant Concentration
During July, August, September 1974

Figure 3



MAX-HOUR OXIDANT CONCENTRATIONS (pphm) 21 June 73
 LOS ANGELES DATA MULTIPLIED $\times 1.4$ FACTOR

Figure 4



MAX-HOUR OXIDANT CONCENTRATIONS (pphm) 27 June 74
 LOS ANGELES DATA MULTIPLIED x 1.4 FACTOR

Figure 5

MAXIMUM 1-HOUR OXIDANT AS A FUNCTION OF NO_x AT VARIOUS NMHC LEVELS

Source: Development of an Oxidant Abatement Strategy
Based on Smog Chamber Data

By: Basil Dimitriadis

August 31, 1973

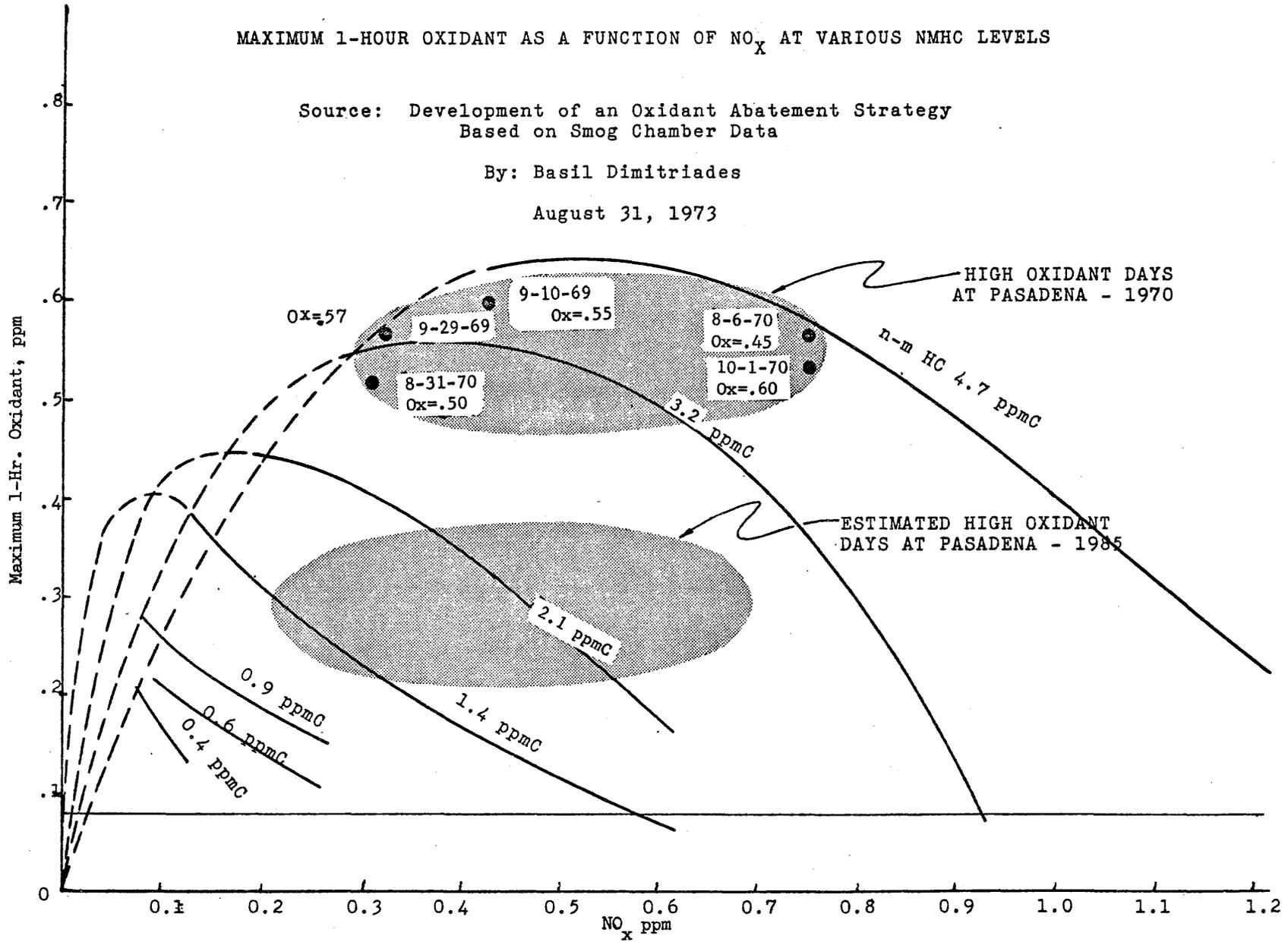


Figure 6

TABLE 1

REACTIVE HYDROCARBONS
ESTIMATED EMISSIONS - LOS ANGELES COUNTY (SCAB)
(TONS/DAY)

YEAR	MOBILE SOURCES	STATIONARY SOURCES	BACKGROUND	TOTAL
1965	896	340	158	1394
1966	873	340	158	1371
1968	806	290	158	1254
1970	753	300	158	1211
1972	644	240	158	1042
1975	479	250	158	887
1980	279	280	158	717
1985	232	310	158	700

SOURCE: ADAPTED FROM STAFF REPORT "EFFECTS OF PROPOSED LIGHT DUTY MOTOR VEHICLE EMISSION STANDARDS ON AIR QUALITY," PRESENTED AT AIR RESOURCES BOARD MEETING, NOV. 13, 1974

TABLE II

OXIDES OF NITROGEN
ESTIMATED EMISSIONS - LOS ANGELES COUNTY SCAB
(TONS/DAY)

YEAR	MOBILE SOURCES	STATIONARY SOURCES	TOTAL
1965	573	320	893
1966	604	310	914
1968	657	290	947
1970	739	260	999
1972	775	240	1015
1975	699	270	969
1980	537	320	857
1985	506	370	876

SOURCE: ADAPTED FROM STAFF REPORT "EFFECTS OF PROPOSED LIGHT DUTY MOTOR VEHICLE EMISSION STANDARDS ON AIR QUALITY," PRESENTED AT AIR RESOURCES BOARD MEETING, NOV. 13, 1974.

TABLE III

HYDROCARBON TO NO_x RATIOS

1B/1B

YEAR	MOBILE SOURCES	TOTAL SOURCES
1965	1.56	1.56
1970	1.02	1.21
1975	.69	.87
1980	.52	.84
1985	.46	.80

$$\text{PPM/PPM} = \left(\frac{1\text{B HC}/14}{1\text{B NO}_x/46} \right)$$

YEAR	MOBILE SOURCES	TOTAL SOURCES
1965	5.13	5.13
1970	3.35	3.98
1975	2.27	2.86
1980	1.70	2.76
1985	1.51	2.63

*OXIDES OF NITROGEN
THREE-YEAR MOVING MEANS OF DAILY
MAX-HOUR CONCENTRATION DURING
JULY - AUGUST - SEPTEMBER*

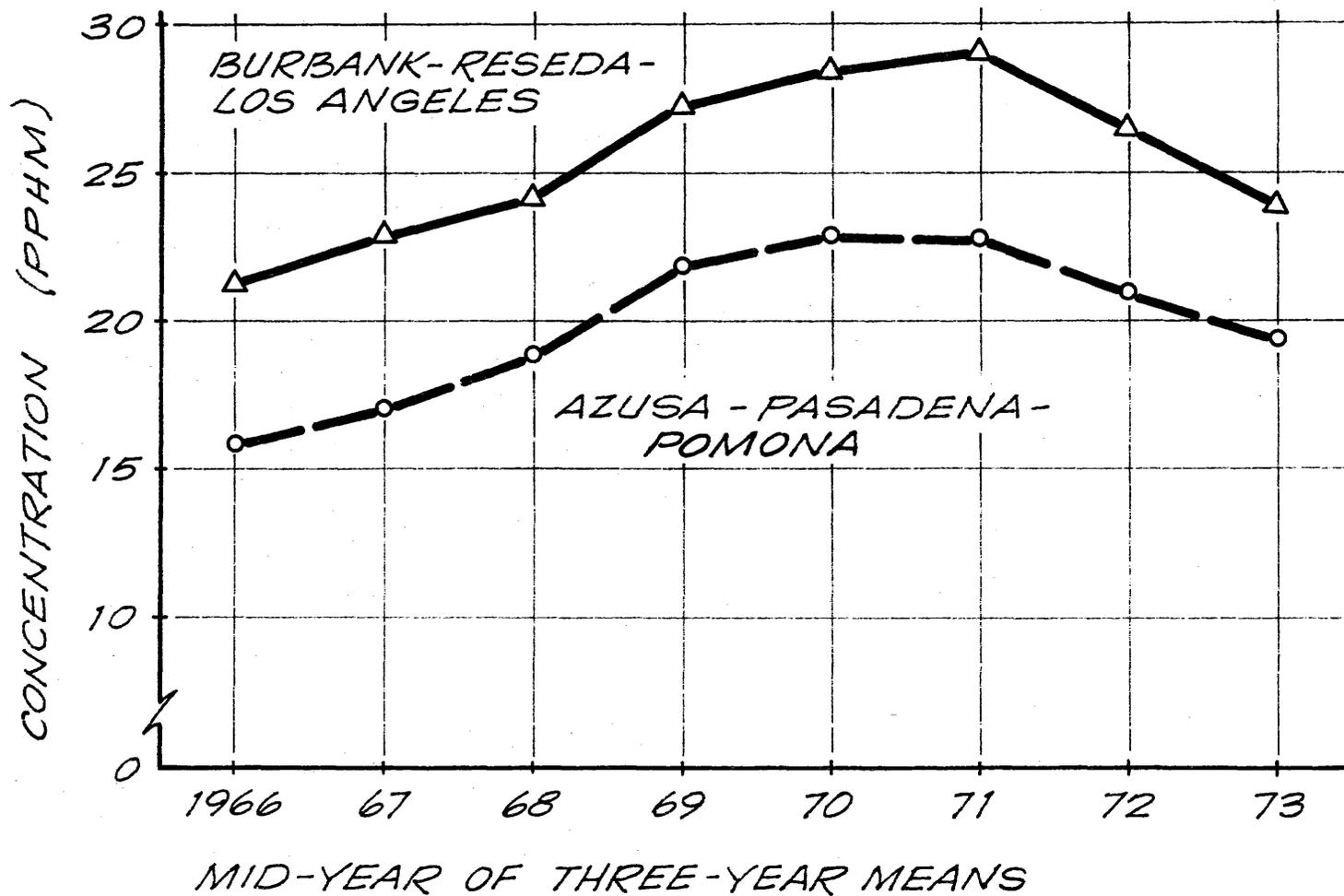


Figure 7

*OXIDANT CONCENTRATIONS
THREE YEAR MOVING MEANS OF
DAILY MAX-HOUR CONCENTRATION DURING
JULY - AUGUST - SEPTEMBER*

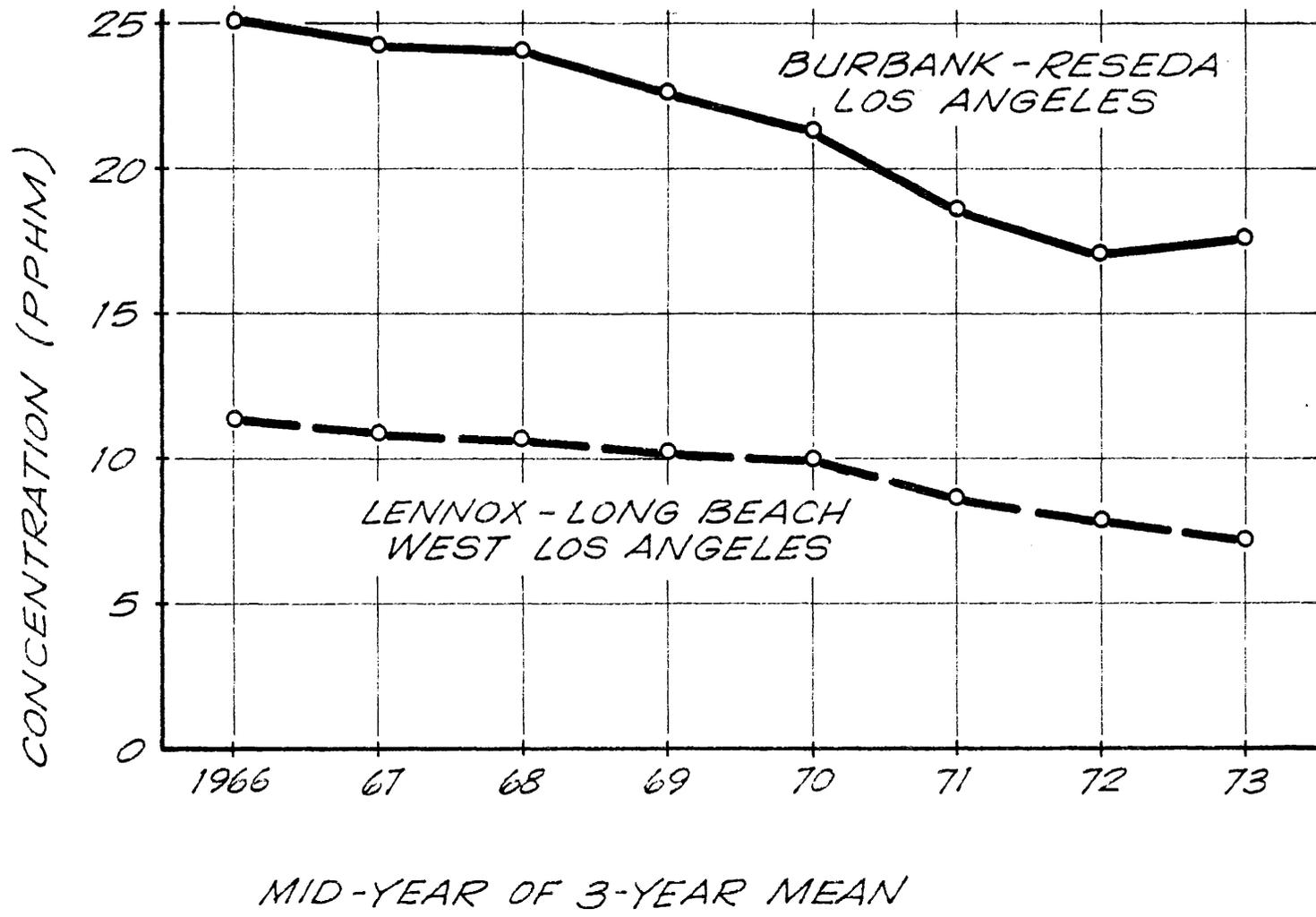


Figure 8

OXIDANT CONCENTRATIONS
 THREE YEAR MOVING MEANS OF
 DAILY MAX-HOUR CONCENTRATIONS DURING
 JULY - AUGUST - SEPTEMBER

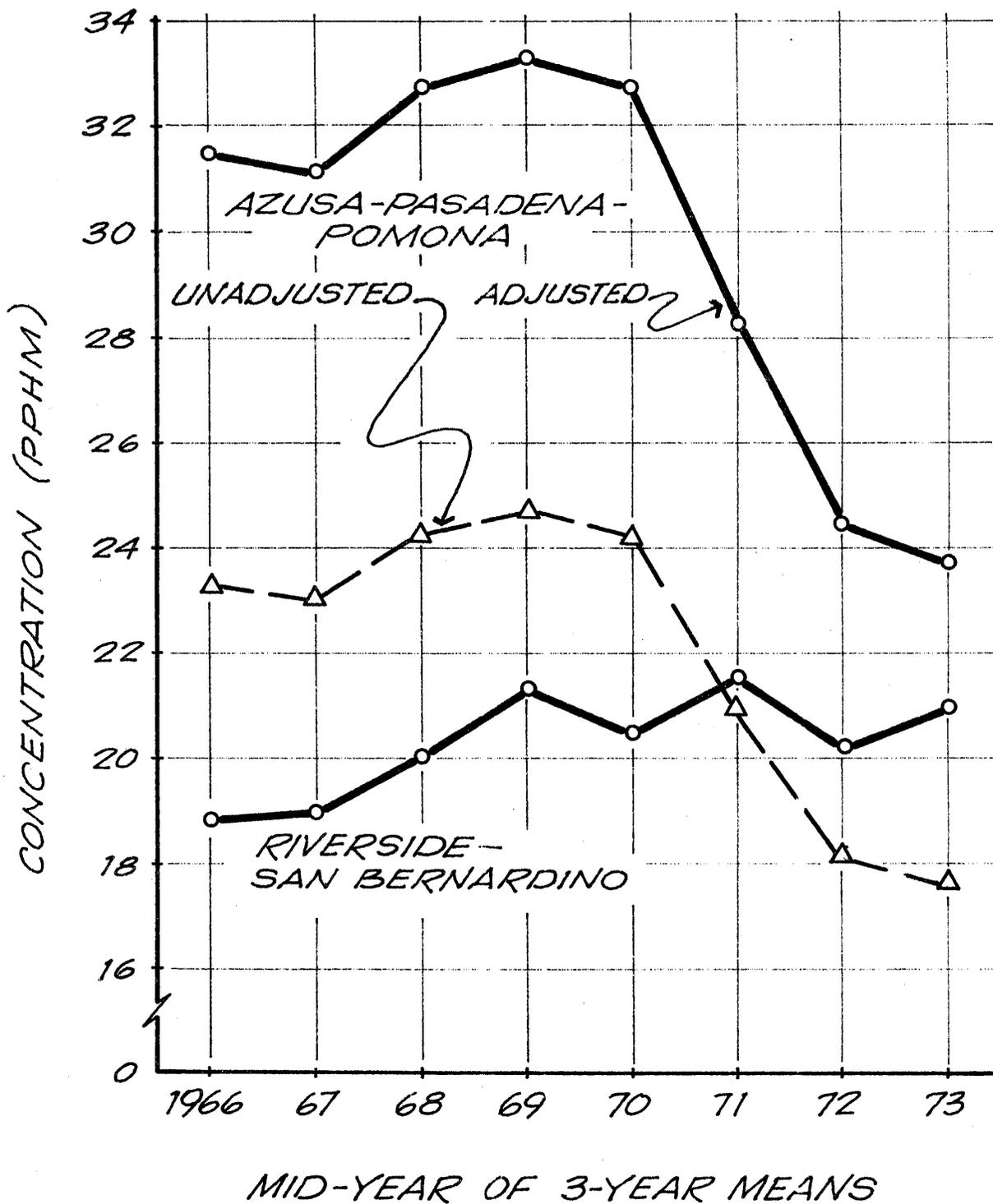


Figure 9

THE USE OF AIRSHED MODELS IN DEVELOPING AND
TESTING CONTROL STRATEGIES

by

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Instead of discussing the use of airshed models in developing and testing control strategies, I will be talking about the fact that they are not being used, and why this is the case.

First, what is an airshed model? The word "airshed" is a very poor name for these models. They are regional air pollution models and we are talking about a model that includes photochemical transformations, emissions of various pollutants that contribute to the reactions, the transport in the atmosphere, and, finally, calculation of ambient concentrations. It is an objective way of translating the emissions through the transport through the reactions to ambient concentrations. Its most useful aspect may be that it is an objective methodology. These regional air pollution models have been developed from two directions: one, from the standpoint of first principles (trying to understand what the actual physical phenomena are, and build upon that and make appropriate approximations); and second, from a statistical standpoint. Both of these end up with some objective technique.

Let's imagine that we have a regional air pollution model that we could use for developing and testing control strategies. What problems would we encounter? First, we would be faced with getting the input data required to run the models; the emissions, the winds, the diffusion, an idea of the reactions, an idea of the insulation (the solar input). We are faced with the

need to compile a tremendous data base in order to run these models. Since we are dealing with a fairly complex model, another problem arises: we can't expect to simulate the effects of a control strategy for every day of the year, for every day of a decade. We must pick out very few days, so we're faced with a decision on what periods to evaluate. Finally, when we are faced with running one of these models, in which we do not have absolute faith, we try to compare the model results to present conditions (i.e., running with present emissions). We have great difficulty in this comparison because the measurements are point measurements and the model predicts something that is not a point value. Even with all these problems, we come back to: if we had a model and it worked it would be an objective methodology.

From a phenomenological point of view, we are faced with using a model that does have a cause-effect pairing, and in areas where we see a cause-effect relationship, we have some sense that we can, either through statistical or first principle approaches, develop a model that may someday agree with reality with sufficient accuracy to use it. But what about areas where we don't necessarily see a cause-effect relationship? If we look at ground-based data (the only data we really have in any detail in areas outside of major source regions), we are faced with looking at the emissions and the air quality and not being able to assign a cause-effect relationship. We are faced with hypothesizing long-range transport aloft. Cases in point: outside the Los Angeles basin are north Los Angeles County and Ventura County, where a local source-local effect relationship is very difficult to draw and we're forced to consider long-range transport.

Another part of the phenomenology of models that can cause difficulty is the emissions inventory. Ozone, or photochemical oxidant, is a time-dependent problem, so we need a time-dependent emissions inventory. We are faced with temporal and spatial averaging for this emissions inventory and with lumping

hydrocarbons into some hydrocarbon classes (e.g., reactive and non-reactive). The emissions inventory problem can introduce major errors. In an emissions inventory we are faced with both point sources and areas sources. Most of the photochemical models do not treat the large point sources very well. This may cause tremendous difficulties in the results. A point source is a very high, localized concentration of a pollutant. However, in most models the atmosphere has been broken up into boxes and, thus, only a number corresponding to the concentration averaged throughout that box is permitted. You cannot represent a point source; it is only done in an average sense, and the averaging may not carry through the non-linear photochemical reactions.

Another part of the phenomenology, one which I think we have under good control at the moment, is the pollutant transport. We have spent many years trying various schemes for simulating transport of pollutants, both wind-blown transport and turbulent transport (the diffusion). Although we have adequate methods, the data to exercise these methods are not forthcoming. Ideally, we would like to have three dimensional winds to know where the pollutants are really going, to know the turbulence, and what is happening to spread the pollutants in three dimensions.

The greatest challenge, however, in photochemical modeling is that of modeling of the transformations, the chemical reactions themselves. We are faced with lumping hydrocarbons in order to get something that can go on a computer and have a reasonable run time. This may be the major problem, but the numerous reactions that can occur is another problem. We are faced with hundreds of possible reactions and reaction steps. Even with all this, we can, under very restrictive conditions after spending a great deal of time, effort, and money, come up with a model that, for these restrictive conditions, can do a good job of simulating.

I would like to describe what we have been able to achieve with two examples. The first is an example of the success that can be achieved with a lot of care in tuning a model and tweeking for a given day. Table I shows the observed and the calculated ozone values for stations in Los Angeles County and Orange County on September 29, 1969. This was actually done by the Systems Applications, Inc., using an Eulerian model. The features of the model are not really important, but the fact is that the results are predictions of the maximum within about an hour of the observance during the day, and within about 10 or 20% of its magnitude. This is the best we can do at this point and the best I think we have reason to hope for at this time in model development. If we extrapolate these results to new conditions, we have great difficulty in knowing exactly how the model will respond compared to reality.

The second example is a graph of the Lagrangian model, DIFKIN, shown in Figure 1. If we start looking at these models in a little more detail, we find that the model does very well during the earlier time period. With the later time period in simulation, the model starts to diverge from the observations. It can diverge significantly from the ambient data, and yet when we are faced with applying the models to an emissions control strategy, we're faced definitely with multiple-day runs.

In addition, some specific problems have arisen in turbulent mixing in which we have to worry whether the constituents actually get in juxtaposition to react. We have to consider aerosol formation, and how that participates in the photochemical reactions. Reaction surfaces may be important and are of concern. The questions brought up earlier in this meeting about whether there exists a definable air parcel, and finally, the multiple-day chemical simulation are very definite problems.

After we have cleared up all these technical problems, we will be faced with the political problems, the problems of developing a model that will

really be used. We are faced with ease of use, cost, and accuracy. Compared to what has been done in the past (rollback, Appendix J, etc.), these photochemical models are not simple to use. The costs are orders of magnitude greater than any other model which has been applied, especially when we are facing multiple day simulations. We want to compare model results to air quality standards. Enforcement people need to be concerned with the absolute numbers, whereas all that the best modelers have been able to hope for is relative numbers, per cent reductions that may be expected.

Finally, we are concerned about the credibility barrier. Even if we have done all our work and we have models that agree with present conditions and they are cheap and easy to use and they have been accurate enough for present conditions, how credible will they be? They will be a complicated black box to the politicians who then have to use the results. These are the present problems, state-of-the-art, and challenges for modeling photochemical, regional air pollution.

Table I

O₃ MAXIMA CALCULATED BY THE EULERIAN MODEL ON 29 SEPTEMBER 1969

Station (California)	Observed Values		Calculated Values	
	Time	Concentration (pphm)	Time	Concentration (pphm)
Reseda	1200	20	1100	22
Burbank	1200	23	1100	23
Pasadena	1100	40	1100	40
Azusa	1300	39	1200	40
El Monte	1200	25	1300	40
Downtown Los Angeles	1000	12	1000	7
West Los Angeles	1100	14	1100	10
Lennox	1100	10	1200	6
Whittier	1100	32	1000	27
La Habra	1200	42	1100	38
Long Beach	1100	7	1300	6
Anaheim	1200	40	1200	38
Santa Ana	1200	21	1200	17

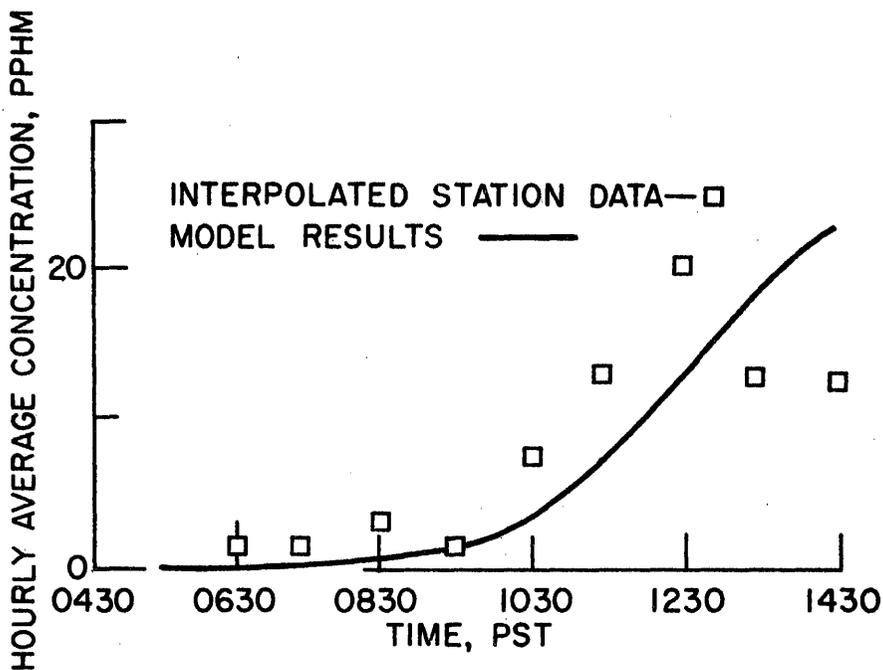
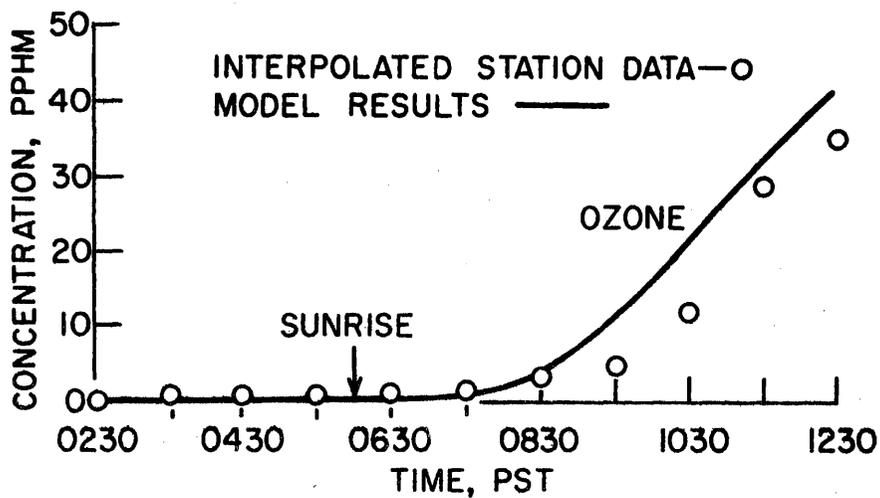


Figure 1. Comparison of observed (O, □) with calculated (—) O_3 concentrations from the Lagrangian model (from Eschenroeder et al.)

THE USE OF AIRSHED MODELS IN DEVELOPING AND
TESTING CONTROL STRATEGIES

by

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It was a bit shocking to see all the preceding speakers giving their fine results and reviewing what they had done because I planned to talk about what we do not know rather than what we do know. So as part of our presentation we rigged it so Ralph Sklarew would view all the problems with alarm and I would point with pride to the solutions.

I will identify the research priorities and some of the outstanding problems as well as suggest some approaches to their solutions for modeling.

Figure 1 lists some of the things which have been mentioned in this meeting one way or the other. Certainly with multiple day simulations, we must be talking about scales that are much larger horizontally and multiple layer vertically if we are going to describe what is really going on in some of the episodes. This also enters the prediction of the impact of high oxidant levels in remote areas where long-range transport is taking place. I will discuss three possible aerosol effects that could be put in models. There are always room for improvements in the air chemistry, and finally, we have to work out some uniform set of standards for assessing how well a model performs.

Figure 2 gives a couple of my notions on this problem of extending the model to a regional scale. I think we must work with a layer that has concentration gradients in it, or with a multiple layer formulation where the mathematical description carries different degrees of precision and different degrees of resolution in the different layers. For example, one can conceive of a model that treats the first 500 meters with a certain vertical mesh size and a certain horizontal cell size and goes up to successive layers into different scales. It would couple the equations from these layers to the bottom one through vertical fluxes induced by the chimney flows that Jim Edinger tells us about. So that we can try to get a simple minded way of transporting the stuff up from the urban surface into these elevated layers where horizontal transport is far more effective than vertical transport. Three dimensional wind fields will be needed for this, but the difficulty can be reduced by holding down the level of detail on this second layer. Thinking about some of the stuff that Neligen was telling us this morning, about these extremely long range transport problems, we may even be able to consider urban areas as point sources and do the reaction within that point source as an initial condition for the long range transport. Then we get into much more complexity in the way of source and sink mechanisms. For example, emissions of reactive hydrocarbons from vegetation, the uptake of ozone on leaf and plant surfaces the adsorption of CO by soils is a very significant feature for long range balance in these processes. Things that we had considered as non-reactive, we now have to consider as reactive. So I think that you can work in this scale on a different level and we hope that its going to be a simpler level than with the faster reactions that we now work with in the urban atmosphere.

Figure 3 shows three aerosol effects that I will discuss here. The first is a chemical process that has to be in the mechanism for producing photochemical aerosol because a great number of the particles that are in the atmosphere were formed by reactions. The second effect occurs once there are particles in the atmosphere. They can act as active sites for catalysis short circuiting certain changes that we otherwise thought had to occur in the gas phase. And, finally, the interaction of aerosol formation in the extinction of ultraviolet light that reaches the surface. Some very interesting radiometry data is coming out from EPA from the LARPP program a couple of summers ago, showing that in the bands where the action is as far as NO_2 dissociation that a significant attenuation can occur. Thus we have a coupled system where the aerosol is being produced by a reaction and is acting to inhibit that reaction at lower levels. So this means mathematically we have to include another equation describing radiant transfer. This describes the extinction of ultraviolet in a series of bands that contribute to the primary photochemical processes. This all sounds discouragingly complicated.

Let us examine one of these aerosol effects in Figure 4. A couple of years ago I did some calculations on the upper limit effect of surfaces. Taking typical urban particular loadings and typical sizes for the particles, one can calculate the area and get a surface-to-volume ratio for the gas parcel considering typical concentrations, as observed for 6 to 9 AM, and then mid-day, one can calculate the ratio of an upper limit (which you get from kinetic theory) of the surface reaction to the gas reaction rate. One can also compute the gas reaction rate. The upper limit assumes that every molecule that hits the particle surface

sticks or reacts thereby losing its identity as soon as it hits. This defines 100% catalytic efficiency, and that is why it defines the upper limit. These results say that a significant fraction of the ozone reaction rate even this early might be due to surface effects. Several more times the gas phase rate is a potential possibility with nitric oxide and nitrogen dioxide, but as Phil Leighton said in his book a long time ago, the oxygen atom really is not going to significantly interact with surfaces. I believe that we can make this kind of generalization for others of the free radicals that are there in very low concentration. Then, the picture somewhat shifts toward ozone being at least potentially a more significant reactor on a surface than the others in the afternoon, and nitrogen dioxide is still hanging in there, but oxygen atom is still many orders of magnitude below where it could even be important. These numbers maybe look large at first glance, but we must remember that the surface reaction efficiency of even ozone, on some organic surfaces are like 10^{-3} , 10^{-4} . Taking this as the upper limit we should really think in terms of perhaps several orders of magnitude below the numbers shown. Perhaps the surface sites on a certain type of particulate form a very active environment for these reactions to take place. So as we get research results from the laboratory on the surface efficiencies we in the modeling trade will be able to take numbers like this and modify them appropriately to decide whether they should be put in the models or not.

As a matter of priorities, I would take item No. 1 and 3 from the list, namely, photochemical aerosol production, and the ultraviolet effect. If we must decide where we are going to put our resources, I believe it should be in the first and third items.

Moving to the question of improvements in air chemistry let us consider Figure 5. I will repeat what several other speakers were saying here. More complicated mechanisms stratify hydrocarbon species into broad classes like parafins, olefins, aromatics, etc., assign these different reactivities and assign these different chain mechanisms. It may be pointless to go to this amount of trouble in chemistry if we do not know how much of these things are being emitted into the atmosphere. If we are really going to be serious about hydrocarbons, we need to know these emissions, broken down by compound class as functions of time and space. The additions of the sulfur compounds in the aerosol formation as mechanisms are really very high in priority.

About three or four years ago I begin warning of the possible perturbation of chemical reaction rates due to inadequate mixing. Some of the chemists charged that this was a figment of the fluid dynamicist's imagination when I first said it, and those same chemists have come back to me more recently saying "Why hasn't something been done about this?" Figure 6 illustrates what started getting us suspicious about it. These data are from 1969, taken by Scott Research Lab at Commerce. What was done here is a plotted point for every ten minutes of data where NO measurements were above 1 part per hundred million. We tested this hypothesis of a photostationary state between ozone, nitric oxide and NO_2 by dividing the forward reaction rate by the reverse reaction rate (which is supposed to be 1, and the log of one is 0 so the photostationary state should be along the axis). Well, naturally because of measurement irregularities you get this big scatter. But it's clear that even with this degree of scatter that we are seeing factor of 10 departures from photostationarity. These are 10 minute averages.

More recent data taken on 6 second time base for LARPP confirms this. The effect is a function of altitude, atmospheric stability, averaging time and of proximity to sources.

What are we going to do about this? It seems to me that we have to devise, first of all, some kind of an empirical correction factor, mainly for O_3 and NO reaction. If we pursued the classical turbulence approach and followed the lead of doing second order closure approximations on all the reactions where this could occur, we would generate over a hundred partial differential equations for even our simple kinetic scheme. This is really out of sight when it comes to computation. I would advocate that the research should concentrate on an empirical correction factor for the NO- O_3 rate at the outset rather than setting forth on a ten year program of turbulence theory. Evaluating or assessing the performance of models, we see the indices heading off in various directions depending on what we want to use the model for. Referring to Figure 7 we note the comparisons between observed and calculated values that are expressed through a correlation coefficient, but this really only measures the model's ability to predict general trends, not necessarily absolute accuracy. This gets us to the questions, "Should we really be asking these models to do specific absolute prediction of levels? Or should we emphasize that they are for relative accuracy?" Most of us prefer when we talk about what's going to happen in 1990 to restrict our attention to relative accuracy. We ask questions about what is the impact of a certain decision on the air quality. We ask whether you make that decision to build something or not to build something, and compare one case with the other, or the impact of that abatement strategy relative to another one.

Finally, there is the acid test, which is a residual error test. This is used if you do want to express absolute accuracy of a model. To do this you must look at the statistics of the difference in between the observed and the calculated and divide it by the observed value.

We are in the process of trying to rationalize the general set of performance indices of models and to look at the performance of specific models as to their impacts on decision making.

Figure 8 sums up a series of questions that I think one should ask before he chooses a model. I think there are a lot of people in audiences like this who are potential model users and should be. We have heard various examples of models from the big box to the big smog chamber and so on. There are several choices that we can make, now. The answer to the first question really hinges around the user, and we don't want to have to have a Ph.D. standing next to a super-computer for all these models. Thus, there should be some element of choice based on who the user is. Very closely related to that is what he wants to use the model for. For example, does he want to give somebody a 90% guarantee that an abatement strategy will achieve the air quality standard? Does he want to assess the impact of building an oil/gas processing plant? Then you must consider the array of models that are available and look at each one's performance indices. It's an "apples and oranges" thing. Some people can make their model look very good by using a correlation coefficient; others prefer to use a residual error analysis so what we really have to do is develop the performance index so that you can use a standardized basis for each model.

Something that is not mentioned very often is, "How readily accessible is the model?" Some models are on computer networks where you can dial

up on a phone and use them. Others are written for a specific computer in a specific language. If you want to transfer it to some other computer, it may be extremely difficult. High degrees of accessibility should be a goal for those of us who are working in this. And finally, I put this at the bottom of the list (but some may wish to put it at the top): the question of cost/effectiveness. I think you can put the cost/effectiveness in perspective by considering the magnitude of the impact of the decisions that are being made. The costs of abatement versus the costs of non-abatement as we see in the economic reports on clean air run into tens of billions of dollars. Thus, spending a few thousand dollars per model run should not really concern us. Nevertheless, it does concern some users of models. They would much rather run the \$100 version than the \$1000 version, but I really believe that when we consider what the models are being used for in terms of the impacts on society and economics we find that this is a relatively minor feature.

In this talk I have tried to bring out some of the problems and the solutions in applying photochemical diffusion modeling to air quality management. We are moving into a new era where the "scalpel" approach must replace the "meat-axe" approach in planning for environmental improvements. Transportation and land-use controls proposed for abatement or maintenance plans must be justified by the best forecasting techniques that are available today.

INCLUSION OF AEROSOL EFFECTS

- PHOTOCHEMICAL AEROSOL PRODUCTION

- SURFACE REACTIONS ON SUSPENDED PARTICLES

- INTERACTION OF AEROSOL FORMATION AND RADIATION
TRANSFER

Figure 3.



UPPER LIMITS ON SURFACE/GAS REACTION RATE RATIOS

<u>SPECIES</u>	<u>6-9 a.m.</u>	<u>MIDDAY</u>
OZONE	8.5×10^{-1}	$4.5 \times 10^{+1}$
NITRIC OXIDE	$2.0 \times 10^{+1}$	6.0×10^{-1}
NITROGEN DIOXIDE	$1.6 \times 10^{+1}$	$1.1 \times 10^{+1}$
OXYGEN ATOM	2.1×10^{-6}	2.0×10^{-6}

Figure 4.



IMPROVEMENTS IN AIR CHEMISTRY
AND EMISSIONS INVENTORIES

- HYDROCARBON EMISSIONS AS FUNCTIONS OF SPACE,
TIME, AND REACTIVITY
- SULFUR/AEROSOL MECHANISMS
- TURBULENT MIXING CORRECTIONS

Figure 5.



POSSIBLE TURBULENT MIXING EFFECTS
OF PHOTOSTATIONARY STATE

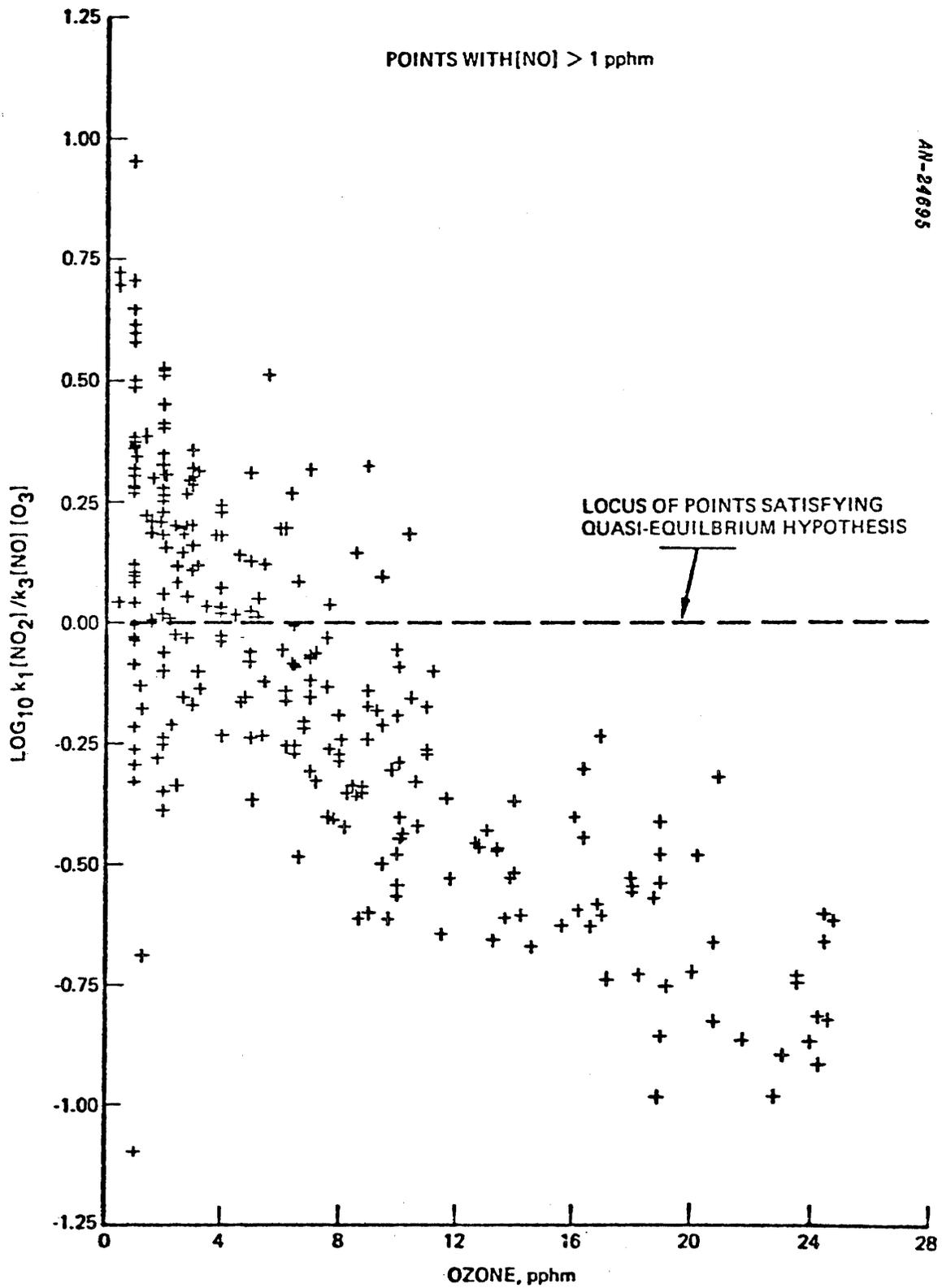


Figure 6.



QUESTIONS ON USE OF MODELS

- WHO WILL BE USING IT?

- WHAT KIND OF QUESTIONS MUST THE MODEL BE EXPECTED TO ANSWER?

- HOW WELL DOES THE MODEL PERFORM FOR THE APPLICATION INTENDED?

- HOW READILY ACCESSIBLE IS THE MODEL?

- WHAT IS THE COST/EFFECTIVENESS OF THE MODEL?

Figure 8.



APPENDIX A

UC-ARB Conference Series Programs, Speakers and
Participants Lists

1. Berkeley, March 18-19, 1974
2. Davis, May 20-21, 1974
3. Riverside, December 16-17, 1974
4. Irvine, March 25, 1975

PROGRAM -- BERKELEY UC-ARB CONFERENCE, MARCH 18-19, 1974

"Chemical and Physical Interactions of SO_x , NO_x , and Photochemical Oxidant:
Current Status and Priorities in Research"

Chairman: James N. Pitts, Jr., Director, Statewide Air Pollution Research Center,
University of California, Riverside

Time	Topic	Speaker
<u>Monday, March 18</u>		
8:45 - 9:00	Welcome and Introductory Remarks	Durward Long and A. H. Batchelder
9:00 - 10:00	Health Effects and Atmospheric Transformations in the SO_x , NO_x , and Photochemical Oxidant System: Research Priorities	John F. Finklea
10:00 - 10:15	Break	
10:15 - 11:00	SO_x from Mobile Sources	Raymond Campion
11:00 - 11:45	NO_x from Stationary and Mobile Sources	Robert F. Sawyer
11:45 - 12:15	Discussion	Richard Perrine
12:15 - 1:15	Hosted Conference Luncheon	
1:30 - 2:15	Physical and Chemical Transformations in Aerosol Systems	George Hidy
2:15 - 3:00	Chemical Kinetics and Mechanisms in the NO_x , HC, and Oxidant System	Harold Johnston
3:00 - 3:15	Break	
3:15 - 4:00	Reactive Intermediates in the NO_x , HC, and Oxidant System and Implications in Modeling Photochemical Smog	Hiromi Niki
4:00 - 5:00	Discussion	James N. Pitts, Jr.

Program, UC-ARB Conference, Berkeley, March 18-19, 1974

Time	Topic	Speaker
<u>Tuesday, March 19</u>		
9:00 - 9:45	Analytical Techniques	Jerome Wesolowski
9:45 - 10:30	Validity of Aerometric Data on Ambient Atmospheres	Milton Feldstein
10:30 - 10:45	Break	
10:45 - 11:30	Chamber Studies in the NO _x , HC, and Oxidant Systems	Edgar Stephens
11:30 - 12:15	Discussion	Peter Mueller
12:15 - 1:30	No-Host Luncheon	
1:30 - 2:15	Chamber Studies in the SO _x , NO _x , and HC System	Arthur Levy
2:15 - 3:00	Application of Aerometric and Chamber Data to Control Strategies	A. Paul Altshuller
3:00 - 4:00	Concluding Remarks and Discussion	John R. Holmes

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PROGRAM - DAVIS UC-ARB CONFERENCE, MAY 20-21, 1974

"Photochemical Smog--Effects and Causes in Agriculture:

Current Status and Priorities in Research"

Regents Room, 203 Mrak Hall

Chairman: O Clifton Taylor, Associate Director, Statewide Air Pollution
Research Center, University of California, Riverside

Time	Topic	Speaker
<u>Monday, May 20</u>		
9:00 - 9:15	Welcome and Introductory Remarks	O. Clifton Taylor
9:15 - 10:15	Priorities for Future Biological Research in Air Pollution	Walter Heck
<u>Effects of Air Pollution on Biological Systems</u>		
10:15 - 11:00	Field Crops	Art Millecan
11:00 - 11:15	Break	
11:15 - 12:00	Biological and Physiological Effects	J. Brian Mudd
12:00 - 12:30	Discussion	Irwin P. Ting
12:30 - 2:00	Hosted Luncheon (Library Room, Faculty Club)	
2:00 - 3:30	Ecosystem	Paul R. Miller
3:30 - 3:45	Break	
3:45 - 4:30	Ornamentals	Richard W. Harris
4:30 - 5:00	Discussion	Joe R. McBride
<u>Tuesday, May 21</u>		
<u>Agricultural Effects on Air Quality</u>		
9:00 - 9:45	Solid Waste	John Goss
9:45 - 10:30	Pesticides, Herbicides, etc.	Donald G. Crosby
10:30 - 10:45	Break	
10:45 - 11:30	Animal Wastes	Ralph E. Luebs
11:30 - 1:00	Summary and Concluding Discussion	Lowell N. Lewis

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"Technical Bases for Control Strategies of Photochemical**Oxidant: Current Status and Priorities in Research"**

Chairman: James N. Pitts, Jr., Director, Statewide Air Pollution Research Center,
University of California, Riverside -- UCR Commons, Large Dining Room

Time	Topic	Speaker
<u>Monday, December 16</u>		
8:45 - 9:00	Welcome and Introductory Remarks	UC and ARB Administrators
<u>Medical Bases for Control Strategies</u>		
9:00 - 10:00	Overview Lecture: Health Effect Implications of Ambient Air Concentrations of Ozone Alone and in Combination with Sulfur Dioxide	David V. Bates
10:00 - 10:15	Break	
10:15 - 11:00	Implications of the Energy Crisis for Oxidant and SO ₂ Control Strategies	Bertram W. Carnow
11:00 - 11:45	Health Effects of Oxidant Exposures: A Research Progress Report	John H. Knelson
11:45 - 12:15	Discussion	Stanley N. Rokaw
12:30 - 1:30	Hosted Conference Luncheon, Faculty Club, UCR	
<u>Technical Bases for Control Strategies</u>		
1:45 - 2:30	Experimental Smog Chamber Studies and Kinetic Computer Modeling in Elucidating Chemical and Physical Transformations in Polluted Atmospheres	James N. Pitts, Jr.
2:30 - 3:15	The Use of Smog Chamber Data in Formulating Oxidant Control Strategies	Basil Dimitriadis
3:15 - 3:30	Break	
3:30 - 4:15	The Use of Ambient Air Data to Develop Control Strategies	A. Paul Altshuller
4:15 - 5:00	Discussion	Frank Bonamassa

Time	Topic	Speaker
<u>Tuesday, December 17</u>		
<u>Automobile Emission Controls: Present, Proposed and Possible</u>		
9:00 - 9:45	The Validity of the Strategy of Linear Rollback of Hydrocarbons to Achieve Oxidant Air Quality Standards	Robert E. Neligan
9:45 - 10:30	Emission Control Devices for Conventional I.C. Engine Designs and Their Implications for Oxidant Control Strategies	Robert F. Sawyer
10:30 - 10:45	Break	
10:45 - 11:30	Comparison of Alternative I.C. Engine Designs for the Control of Automobile Emissions	Henry Newhall
11:30 - 12:00	Discussion	Rhoads Stephenson
12:00 - 1:15	No-Host Luncheon	
<u>Emission Inventories, Transport and Models in Control Strategy Formulation</u>		
1:15 - 1:45	Airborne Surveys in Urban Air Basins: Implications for Model Development	Donald Blumenthal
1:45 - 2:15	Relative Contributions of Stationary and Mobile Sources to Oxidant Formation in Los Angeles County: Implications for Control Strategies	Robert G. Lunche
2:15 - 2:45	Oxidant Measurements in the South Coast Air Basin: Implications for Control Strategies	John R. Kinosian
2:45 - 3:00	Break	
3:00 - 3:20	The Use of Airshed Models in Developing and Testing Control Strategies	Alan Eschenroeder and
3:20 - 3:40		Ralph Sklarew
3:40 - 4:30	Concluding Remarks and Discussion	Milton Feldstein

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CONFERENCEHealth Surveillance Related to Air Pollution

March 25, 1975 - 8:30 a.m. to 5:00 p.m.
Holiday Inn, Citation Room
3131 Bristol, Costa Mesa, California
Telephone: (714) 557-3000

Co-Sponsors: Air Quality Advisory Committee, California State Department of
Public Health John Goldsmith, M.D., Chairman

University of California-Air Resources Board Conference Contract
James N. Pitts, Jr., Ph.D., Contractor and General Chairman
T. Timothy Crocker, M.D., Conference Convenor

Dear Colleague:

A conference has been planned which will extend a meeting of the Air Quality Advisory Committee of the California State Department of Health into a University of California-Air Resources Board "mini-conference" on air pollution. This conference is the fourth in a series being conducted by the Statewide Air Pollution Research Center for the Air Resources Board. The Committee will convene at 4:00 p.m. on March 24 at the Holiday Inn, Costa Mesa, near the Orange County Airport.

You are invited to participate in the conference which will occur from 8:30 a.m. to 5:00 p.m. on March 25 in the same location. The advantage of this joint session is to coordinate the planning efforts of the Health Department, ARB and UC to identify the elements required for a health surveillance program related to air pollution. The scale of the program may be small but the scope should be comprehensive. Some relevant topics were selected and speakers for each topic were invited to present data and/or describe resources suitable to a health surveillance program. A number of qualified participants have also been invited to be present without being asked to prepare a statement. The object will be to engage interested investigators and institutions within the State in planning and presenting an integrated proposal for an air pollution health effects surveillance program.

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Enclosures

PROGRAM

8:30-8:35 a.m. Introduction of Conference objective
T. T. Crocker

8:35-8:45 Statement of interests of Department of Public Health
J. Goldsmith

8:45-8:55 Function of Legislative Technical Advisory Committee
J. N. Pitts, Jr.

Session I. Air Pollution Patterns in the South Coast Air Basin

8:55-9:15 Monitoring networks and capabilities in the South Coast
Air Basin
J. Kinosian, J. Paskind

9:15-9:25 Discussion

9:25-9:45 Monitoring for aerosols by size and composition
T. Cahill

9:45-9:55 Discussion:
J. Wesolowski

9:55-10:05 CHESS station distribution and monitoring capability
C. R. Thompson

10:05-10:20 Coffee

10:20-10:50 Air pollution "corridors" and micrometeorology of the
Air Basin including localization of areas of sulfate,
nitrate and oxidant pollutants
W. White

10:50-11:10 Discussion:
W. Kuby

11:10-11:20 Technologic needs in micrometeorology, monitoring and
pollutant characterization
P. Mueller

11:20-11:50 Discussion

12:00-1:30 Luncheon

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Conference on Health Surveillance Related to Air Pollution

March 25, 1975
Costa Mesa, California
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APPENDIX B**Abstracts of papers presented at UC-ARB Conferences**

1. Berkeley, March 18-19, 1974
2. Davis, May 20-21, 1974

Abstracts for Berkeley Conference

NO_x FROM STATIONARY AND MOBILE SOURCES

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A number of basic questions are important to assessing the nature and control of sources of the oxides of nitrogen. Some of these questions have been answered, some not. The accuracy of some of the answers remains in doubt.

1) What is the form of the oxides of nitrogen? NO_x refers to a mixture of NO and NO₂. Source nitrates have not been identified as a major effluent, probably accurately but no major attempts to identify source nitrates have been publicized. It has been generally thought that combustion sources of NO_x were predominately NO, usually the figure greater than 95% is used. More recent studies suggest that NO₂ may be the dominant species from some sources. How significant conversion from NO to NO₂ might take place in the exhaust systems of typical sources is not understood.

2) Where do the oxides of nitrogen come from? Non-combustion sources, such as fertilizer and explosives plants are known to be important point sources. Of greater general importance are the combustion sources--these account for about 99% of the oxides of nitrogen. Nationwide combustion sources are about equally split between stational and mobile categories--in local regions substantial variations exist. Generally accepted source estimates probably underestimate refinery contributions.

3) How are the oxides of nitrogen formed? In combustion processes two major routes which form nitric oxide are important. "Thermal" nitric oxide results from the fixation of molecular atmospheric nitrogen in the high temperature combustion process. "Fuel nitrogen" nitric oxide results from the selective oxidation of nitrogen bound in the fuel--high temperatures are not essential. The fuel nitrogen process is not well understood and is of increasing importance as coal and synthetic crudes from oil shale and coal become more important energy sources. Closely related is the ill defined but observed occurrence of reduced nitrogen compounds in combustion products, e.g., NH₃ and HCN.

4) What is the relationship between the source and its impact on air pollution? Large point sources of oxides of nitrogen may present excessive downwind exposures. Plume conversion of nitric oxide to nitrogen dioxide including the problem of an oxidizing atmosphere is not well understood. Distributed sources, such as automobiles and home appliances may have a quite different air pollution impact, especially if NO_x is emitted into "developed smog". High altitude

emissions of the oxides of nitrogen from aircraft pose a special threat. The importance of exhaust plume reaction is only partially resolved.

5) How are the oxides of nitrogen controlled? What are the costs of control? Thermal nitric oxide is most directly controlled by limiting maximum combustion temperature. Although this is often difficult in practice, there are not fundamental reasons why efficient combustion cannot be achieved at temperature below which nitric oxide is formed. Current approaches to nitric oxide controls from automobiles appear to carry fuel economy penalties if carried to the .4 gm/mile level. Nitric oxide control methods without fuel economy penalties need to be developed. Note that if a truly effective NO_x catalyst were to exist then the engine designer would be free to achieve greater engine efficiency through the traditional approach of higher compression ratio and optimum spark timing. Considering the cost and difficulty of attaining 90% reduction in automotive NO_x emissions, greater attention should be paid to the reduction of stationary NO_x sources. Combustion modification for control of power plant thermal NO_x is effective and generally improves plant efficiency. The fuel nitrogen control problem is much more difficult and promises to become an increasing problem on an immediate time scale with conversion to oil and coal fuels. Control related problems remaining to be solved range from fundamental chemical kinetics to large power systems design. Fuel modification to remove fuel nitrogen may be required.

The major unresolved problems related to NO_x from stationary and mobile sources are focused in the understanding and control of the combustion processes in which the oxides of nitrogen are formed. These problems are likely to increase as shortages bring about a shift to fuels containing higher concentrations of fuel nitrogen. The natural tendency of higher performance combustion devices has been toward the production of higher levels of the oxides of nitrogen--there is no fundamental reason why this must be so.

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Abstract for ARB/SAPRC Research Conference, Berkeley, CA, March 18-19, 1974

PHYSICAL AND CHEMICAL TRANSFORMATIONS IN AEROSOL SYSTEMS

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ABSTRACT

This paper presents a brief review of recent progress in characterizing aerosol behavior in the presence of photochemical smog. Recent field studies led by the California Aerosol Characterization Experiment (ACHEX) have provided an extensive data base for defining the important contributors to photochemical aerosol evolution. Data taken in the South Coast Basin indicate that more than half of the aerosol collected is material produced in the atmosphere and not from primary sources. The bulk of the photochemical aerosol is made up of sulfate, nitrate, non-carbonate carbon, and liquid water.

Recent measurements from ACHEX show distinct and important relationships between the diurnal behavior of certain trace gases such as O_3 and NO_2 and the constituents $SO_4^{=}$, NO_3^- , and carbon. These data are crucial to developing an improved picture of the mechanisms of photochemical processes and aerosol behavior. Selected observations are used to illustrate the correlation between $SO_4^{=}$, carbon, and ozone as contrasted to the patterns of correlation between NO_3^- and NO_x .

The ACHEX data further show major differences in the distribution of $SO_4^{=}$, C, and NO_3^- with respect to size in the airborne particles. Examples of these variations indicate that the former dominate the particles $< 0.6 \mu m$ diameter, while the latter is confined largely to the $> 0.6 \mu m$ range.

Recent laboratory experiments tend to verify conclusions of early work on the formation rates of particles for SO_x , air, X mixtures. New programs on irradiated NO_x , He, air systems and O_3 -HC systems show promise for improving the knowledge of the significant aerosol forming reactions in the atmosphere.

Listed by priority are several important research questions on photochemical aerosols:

1. What are the growth rates for photochemical haze?
2. What are the hydrocarbon vapor species that are involved in aerosol formation - are they similar in reactivity scale to those for oxidant formation?
3. Are reactions dominated by homogeneous or heterogeneous processes?
4. Are the constituents SO_x , NO_x , and non-carbonate carbon synergistically involved in aerosol evolution?
5. What role does liquid water play?
6. What are the NO_x species responsible for NO_3^- production?

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ABSTRACT

Despite inherent complexities arising from the presence of a multitude of reactive chemical species in urban atmosphere, significant progress has been made over the years in the understanding of smog chemistry. In particular, experimental and theoretical studies of NO_x-HC-Oxidant systems and relevant elementary reactions^{1,2,3} have led to the formulation of various reaction schemes now in use together with airshed models for the assessment of urban air quality.^{4,5,6} Illustrated in Table 1 is a state-of-art "lumped" chemical model which is compatible with existing aerometric data. In this table, R denotes H atom or alkyl radicals in a lumped form. These reactions represent a minimal set of crucial chain oxidation steps of smog reactions triggered by photodissociation of NO₂, nitrites, nitrates, aldehydes, and peroxides. Concentration-time profiles of NO_x, HC, oxidants, and various other reaction products are greatly affected by the reactive intermediates which comprise a variety of oxygenated radicals such as NO₃, N₂O₅, RO, RO₂ and R(CO)O₂. Kinetic features of NO_x-HC-oxidants will be presented briefly with specific reference to these reactive intermediates.

Numerous uncertainties still exist in the kinetics and mechanisms of the elementary reactions given in Table 1.^{2,3,7,8} More importantly, no adequate sets of smog chamber data are currently available for the validation of smog models.^{2,3,4} Concerted efforts in the following areas of laboratory work appears to be of particular importance in future developments of models for atmospheric chemistry.

Needed Information on Elementary Reactions:

Many of these smog reactions are not amenable at present for direct studies because of experimental difficulties associated with generation and detection of these polyatomic radical species. Kinetic studies of relatively simple multi-component reaction systems appear to be a plausible approach for the immediate future. In addition, some crucial reactions may still be overlooked. Some research effort should be spent on a systematic examination of the entire matrix of thermochemically feasible reactions. Indicated below are some of the most urgently needed information on the elementary reactions.

- . Hydrocarbon oxidation reactions (Reaction 10-17 in Table 1): particularly, mechanisms of O₃-and OH-reactions of olefins and aromatics, and photolysis of aldehydes.
- . Competitive reactions of oxygenated radicals with NO, NO₂, O₂ and themselves. (Reactions 18-29)
- . Chemistry of Nitrous Acid (Reactions 8-9)

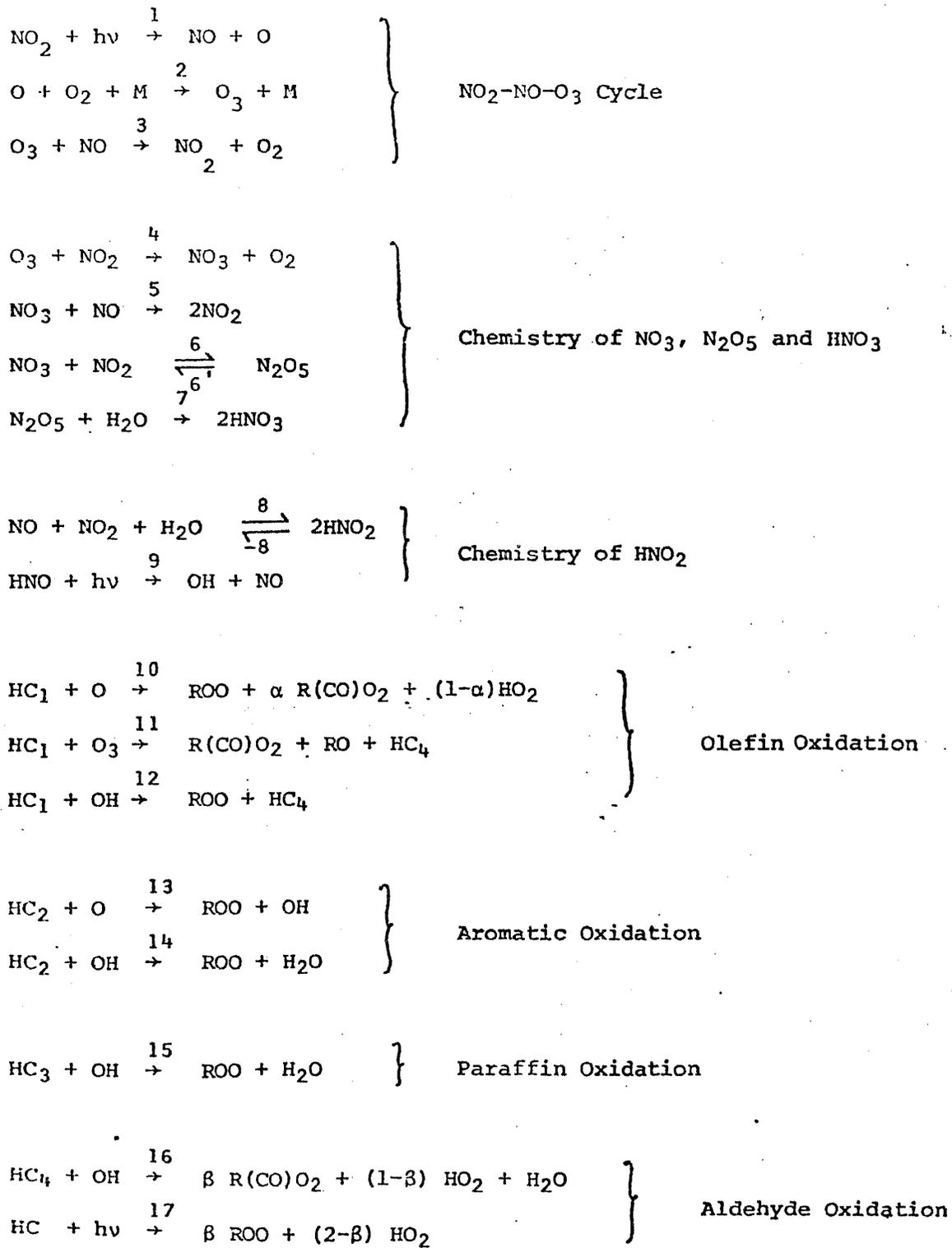
Critical Factors in Smog Chamber Studies:

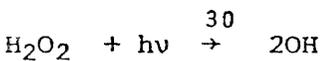
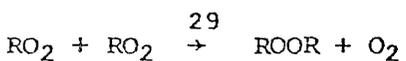
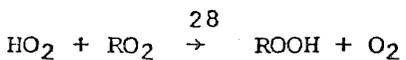
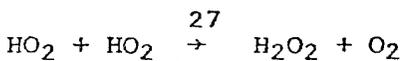
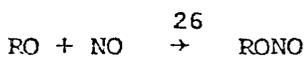
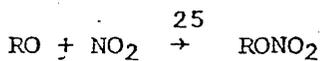
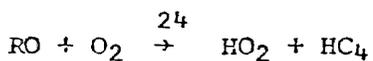
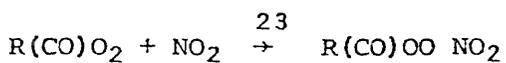
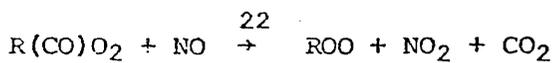
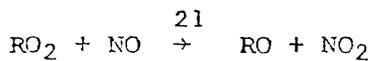
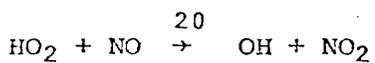
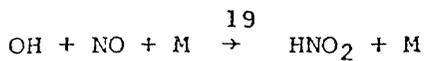
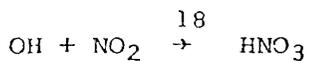
Although existing smog chamber data have played a vital role in the development of smog models, many of the crucial parameters of these experiments are neither well defined nor controlled adequately. Future photochemical studies of NO_x-HC-Oxidant system designed for validation of smog models should be conducted with particular attention to the following experimental factors. It would appear that an evacuable cylindrical reactor equipped with an end-on illumination and with Fourier Transform i.r. spectrometer⁹ provides desirable monitoring and control for many of these factors.

- . Irradiation Source: homogeneity, spectral distribution and intensity, and chemical and physical monitoring.
- . Chamber Design and Operation: Sample preparation and mixing, sample volume for analyses, cleaning procedure, absorption-desorption characteristics of reactor walls, S/V, temperature and humidity control.
- . Analytical Techniques: Universal in-situ fast response, high intensity, selective detection method. Detection of reactive intermediates such as OH and NO₃.

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A Typical Photochemical Smog Model (Ref. Seinfeld et al.)



Reactions of
Oxygenated
Radicals with
NO, NO₂, and O₂

Peroxy Radical
Recombination Reactions

Photolysis of H₂O₂

ABSTRACT

CURRENT STATUS AND PRIORITIES OF
ANALYTICAL TECHNIQUE DEVELOPMENT☆

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This talk will delineate the current status and priorities of Analytical Technique Development for Urban Smog research. We will discuss what pollutants are normally measured in urban smog research programs, what specificities, sensitivities, accuracies, etc. are needed for these measurements in order to reach the specific objectives of the programs, what is the current status of the techniques needed for this research, and what are the priorities for future analytical technique development efforts.

During the presentation a number of recommendations will be made regarding the general area of analytical techniques development. Some of these are based on the data listed in Tables 1 and 2 which are attached.

The following recommendations are reproduced here in order that the participants may have an opportunity to read them before the meeting thus allowing for more active participation during the discussion periods. They are not necessarily listed in order of priority.

☆Submitted for presentation at "Chemical and Physical Interactions of SO_x, NO_x, and Photochemical Oxidant: Current Status and Priorities in Research", Berkeley UC-ARB Conference, March 18-19, 1974.

1. More effort should be placed on determining the equivalency and accuracy of methods, particularly with respect to the chemistry of the aerosol. Specifically the equivalency of the more common methods presently in use for the determination of sulfur compounds in ambient air aerosol should be determined. Table 3 categorizes some of these methods. "Direct" refers to methods which require little manipulation of the sample whereas "indirect" refers to wet chemical methods.
2. Improvements are needed in the methods for the measurement of gaseous NH_3 and specific hydrocarbons.
3. Methodology must be developed for sulfuric acid mist, Pt and Pd.
4. Improvements are needed in the methods for $\text{SO}_4^{=}$, NO_3^- , NH_4^+ , specific particulate carbon compounds, and H_2O .
5. It would be useful to determine the degree of equivalence between the determination of total sulfur in ambient air aerosol as measured by a simple rapid method such as x-ray fluorescence analysis and water soluble sulfate as obtained by the more time consuming wet chemistry.
6. More effort should be placed on determining the quantitative and qualitative usefulness of ESCA for aerosol characterization.
7. High priority should be given to the development of a method which semi-continuously determines the sulfur content of aerosol at nanogram levels. One such system that shows promise is the thermal-flame photometric detector system.
8. There should be further work to determine the usefulness of the thermal-flame ionization detector combination which obtains the concentration of carbon species in aerosol as a function of their volatility.

9. There is a need to develop a reference multi-stage aerosol impactor, that is, one which has negligible bounce off and wall loss and for which the effective cut-off diameters are well known.
10. In many cases the largest error in the determination of the chemistry of the aerosol is the sampling error, not the analytical error. Therefore, for those agencies responsible for funding research on improvements in methodology more emphasis should be placed on improvements in aerosol sampling.
11. More emphasis should be placed on research to develop substrates for collecting aerosol that minimize both the analytical and the sampling errors.
12. High priority should be given to develop a size segregating sampler for aerosols of diameters below 0.5 microns which provides sufficient sample for chemical analysis.
13. Development of "in situ" methods for chemical analysis of aerosols should be emphasized since errors associated with the collection of the aerosol play such a dominant role.
14. To better understand aerosol formation mechanisms it is important to determine the spatial distribution of chemical species in individual particles. Therefore, development of such physical methods as the ion microprobe should be conducted.
15. The quantitative value of elastic scattering of ions (e.g. alpha particles) for determining light elements such as carbon, oxygen, and nitrogen in aerosol should be studied.

16. The quality rather than the quantity of the data should be stressed. Thus it is recommended that atmospheric research proposals not be considered for funding unless they contain a quality assurance section which clearly delineates what tests and procedures will be undertaken to assure the funding agency of the quality of the data obtained. Approximately one out of every five dollars spent on atmospheric research will have to be used to carry out the necessary quality assurance effort. It is simply not true that bad data is better than no data at all.

TABLE I

BASIC CONTINUOUS GAS MEASUREMENTS
FOR URBAN SMOG RESEARCH

<u>Gas</u>	<u>Detection Limit Required</u> (ppm)	<u>Present Capability</u>		<u>Technique* Development Priority</u>
		<u>Technique</u>	<u>Detection Limit</u> (ppm)	
SO ₂	0.005	FPD	0.005	low
O ₃	0.005	Chemiluminescence	0.005	low
NO	0.005	Chemiluminescence	0.001	low
NO ₂	0.005	Chemiluminescence	0.001	low
CO	0.1	Catalytic Reduction/FID	0.1	low
NH ₃	0.001	Chemiluminescence	0.001	high
		Microwave Resonance	0.001 (?)	
		H ₂ SO ₄ Bubbler	0.005	
Specific HC	0.001	GC/FID	0.01	high

*This estimated priority is based on the sensitivity, accuracy, and specificity required.

TABLE II

BASIC AEROSOL MEASUREMENTS
SEMICONTINUOUS AS A FUNCTION OF PARTICLE SIZE
FOR URBAN SMOG RESEARCH

<u>Aerosol</u>	<u>Detection Limit Required</u> (μg)	<u>Present Capability</u>		<u>Technique* Development Priority</u>
		<u>Technique</u>	<u>Detection Limit</u> (μg)	
H ₂ SO ₄	0.01	Humidified nephelometer Microdiffusion (Monkman)	-- 10	high
SO ₄ ⁼	0.01	ESCA AIHL Micro-technique	0.003 0.2	high
NO ₃ ⁻	1	Micro-xyleneol	1	moderate
NH ₄ ⁺	1	Indophenol-blue	10	high
Total Sulfur	0.01	FPD, XRFA	0.01, 0.3	high
Carbonaceous material	0.001-1	ESCA Thermal-FID Mass Spec	0.003 1 0.001	high
Pb, Br	1	XRFA	0.005	low
Na, Cl, Al	1	NAA	0.001	low
V	0.01	XRFA, NAA	0.02, 0.001	moderate
Pt, Pd	0.01	AAS	20, 3	high
H ₂ O	5	Microwave waterometer	5	high
Mass	100	B-gauge Gravimetric	10 25	moderate

*The estimated priority is based on the sensitivity, accuracy, and specificity required.

VALIDITY OF AEROMETRIC DATA
IN AMBIENT ATMOSPHERES
AND THEIR APPLICATION TO CONTROL STRATEGIES

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ABSTRACT

Control strategies have been developed for the reduction of reactive hydrocarbon emissions required to meet the Federal Ambient Air Quality Standard for Oxidant of 0.08 ppm. These strategies are based on calculations relating maximum oxidant levels reached in an air basin during a base year and a linear roll back model to calculate the allowable emission of reactive hydrocarbons required to meet the standard. The results of these calculations indicated a needed 78% reduction in hydrocarbon emissions in the Bay Area if the standard is to be achieved by the 1977 deadline. These programs, if implemented, will impose a severe burden on the people of the Bay Area. Before embarking on such programs, there should be searching inquiries into the scientific validity of the assumptions made - such questions as:

1. What is the reactivity of classes of organic compounds to be controlled and what is their participation in oxidant formation in the real world atmosphere?
2. What is the validity of using single high hour oxidant value, occurring at a single point in a 6,000 square mile area, to calculate the needed reduction for the whole area?
3. What is the validity of using a linear roll back model to estimate required organic compound reduction?
4. How accurate are our estimates on actual emissions of reactive organic compounds from mobile and stationary sources, and how do these estimates affect the calculations used for roll back?
5. What is the role of natural background ozone relative to the standard?

These and other questions must be answered before we embark on programs which would seriously affect every resident in the urban areas of California.

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CHAMBER STUDIES IN THE NO_x, HC AND OXIDANT SYSTEMS

Edgar R. Stephens

Abstract for the Berkeley UC-ARB Conference, March 18-19, 1974

In the quarter century in which laboratory chambers have been used to study photochemical smog, the research questions have become progressively more demanding. The first projects only attempted to establish whether smog symptoms could be qualitatively reproduced in a laboratory environment, and if so, what primary pollutants were necessary. Other early studies were concerned with the feasibility of smog abatement through control of fuel composition. Later studies, such as those at the public health service, were aimed at assessment of the health hazards of this new kind of air pollution. Since control of automotive exhaust emissions has proven to be both difficult and expensive, much recent chamber work has been directed toward development of emission standards for autos. This has proven to be a complex problem because it is now clear that the dependence of oxidant (ozone) and other toxic reaction variables is far from simple. It will be argued that laboratory smog chamber studies can make an important contribution to the control strategy and standard setting effort. But it will be necessary to bridge or at least narrow the gap between ambient polluted air and laboratory conditions.

The laboratory system must be capable of generating a quantitatively realistic smog from reactant concentrations, light intensities, and irradiation times that are also realistic. We now have sufficient analytical data on real polluted air to make detailed comparisons possible, not only for oxidant (ozone), PAN and other reaction products, but also for degree of reaction as judged by the altered hydrocarbon distribution of reacted mixtures. This should make it possible to narrow considerably the gap between chamber data and ambient air data. If this gap can be bridged and chamber data reconciled

with polluted air data then control strategy could be soundly based on both. The second gap which needs closing is between ambient air and chamber data on the one hand, and emission standards on the other hand. The first step is to coordinate the methods of measurement. Much chamber data on hydrocarbon reactivity for example, has been expressed on a ppm basis whereas emission standards are invariably expressed on a weight basis. Hydrocarbon reactivities will have different relative values if compared at equivalent micrograms per cubic meter rather than at equivalent ppm. The hydrocarbons of low reactivities are principally affected.

Still lacking from chamber study literature is a good simulation of photochemical aerosol formation. This is becoming more important because of the evidence that particulate, in addition to impairing visibility, also influences health. Even though many laboratories have reported aerosol formation in synthetic systems, none has yet succeeded (or even seriously attempted) to synthesize and study a truly realistic aerosol formed under truly realistic conditions. This would be of immense help in interpreting the data now being accumulated on ambient aerosols.

The emphasis should be toward closing the gap between chamber studies, ambient air studies, and emission standards. All three efforts will benefit from a successive attempt to correlate data.

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BERKELEY UC-ARB CONFERENCE

on

CHEMICAL AND PHYSICAL INTERACTIONS OF SO_x , NO_x , AND
PHOTOCHEMICAL OXIDANT:
CURRENT STATUS AND PRIORITIES IN RESEARCHMarriott Inn
Berkeley, California

March 18-19, 1974

Chamber Studies in SO_x , NO_x , and HC SystemsArthur Levy
Battelle, Columbus Laboratories

This discussion will address itself principally to the effects of SO_2 additions to simulated polluted atmosphere reactions in environmental chambers. As such, past work and to a large extent current work is concerned with the homogeneous gas-phase oxidation of SO_2 in hydrocarbon (HC), i.e., all organics- NO_x systems. At the outset it is important not to lose sight of the fact that SO_2 removal processes, as we understand them today, are governed principally by heterogeneous processes. However, we also see strong evidence for important homogeneous SO_2 interactions in photochemical smog systems, principally via chamber-type studies. And it is in this regard that we examine this subject.

The principal questions we are addressing ourselves to in the discussion of chamber experiments are: (1) what is the upper limit to the homogeneous rate of photooxidation of SO_2 to SO_3 ? (2) what is the mechanism of the process? and (3) how does the homogeneous process bear on the nucleation and growth of aerosol in the atmosphere?

To maintain perspective to the importance of heterogeneous removal processes a short resume of the current state of knowledge in heterogeneous removal studies is presented. Principally we note here the variability of SO_2 half-lives in atmospheric plume studies and the variability of oxidation rates in solution oxidation studies (laboratory and

theoretical). These studies suggest that heterogeneous SO_2 removal processes may be as low as 30 percent/hr but probably are more like 50 percent/hr in most instances.

Chamber studies in the past have been mainly concerned with understanding, or possibly more correctly stated, observing the effects of adding SO_2 to photochemical smog systems, i.e., irradiated systems of SO_2 -HC-NO- NO_2 -air. Increases in aerosol formation were noted; effects on ozone (oxidant) formation were noted--in the main, the older studies were principally phenomenological in character, not analytical. Currently, in the past few years, the trend has been to examine the role of SO_2 more critically. Some remarks are therefore addressed at the outset of this phase of the discussion to the more fundamental aspects of the photo-oxidation of SO_2 . Thus, on the basis of various studies with hydrocarbons, oxygen, and SO_2 where quantum yields of 10^{-2} were observed, it would appear that one percent of the excited SO_2 is in the triplet state, $^3\text{SO}_2$. Calvert's group in turn has observed high quenching rates for olefin hydrocarbons, suggesting that one or two excited SO_2 molecules out of 10^4 can react with olefin; this further suggests that the marked influence of SO_2 on aerosol formation in smog processes may be due in part to the olefin quenching reaction.

Theoretical rates of homogeneous oxidation of SO_2 estimated from these quenching considerations suggest a maximum rate of 2 percent/hr. Other considerations, based on potential homogeneous reaction paths, suggest a removal rate of almost 5 percent/hr. Recent chamber experiments, designed with more care to answering the rate of removal question, suggest values from 4-10 percent/hr. In these experiments SO_2 is photooxidized in clean air systems and in simulated polluted air systems. Good agreement is obtained on comparing rates of SO_2 removal and rates of sulfuric acid formation.

Recent smog chamber studies on nucleation and aerosol growth processes in homogeneous SO_2 -HC- NO_x systems are presented. Electrical mobility analyzer data on acid aerosol growth are also related to SO_2 removal rates in these studies. Observations are also presented on HC- SO_2 interactions in aerosol formation which may help elucidate mechanistic paths of SO_2 removal in homogeneous smog systems. It is noted for example that aerosol from a SO_2 -air system will appear as soon as the irradiation is initiated, will be delayed slightly in a HC/ SO_2 / NO_x system, and will be

further delayed in a HC/NO_x containing no SO_x.

Attempts to fit mechanisms to smog chamber results remain quite academic in the area of SO₂-smog interactions. A typical example of some of the complexities relative to the influence of SO₂ on ozone production in simulated polluted atmospheres follows. In the 1-butene system under certain conditions of humidity and HC/NO_x ratio ozone formation is reduced in the presence of SO₂. In the toluene system ozone production is increased. In some recent studies in SO₂-clean air systems on the other hand near agreement in the production of ozone and sulfuric acid support an SO₄ mechanism. Much work is needed to bear out these incidental areas of agreement or disagreement in mechanisms.

In summary, we still find ourselves in a quandary as to how meaningful the homogeneous removal of SO₂ in the atmosphere is relative to heterogeneous processes. The latest evidence of rates of 4 percent/hr, possibly as high as 7-10 percent/hr, although low relative to heterogeneous removal processes cannot be set aside as too low for concern. With the rather limited work done to date the evidence still suggests that SO₂ may be removed at significant rates in irradiated HC-NO_x systems. The important question remains, what is the maximum homogeneous removal rate? To finally establish this point more information is needed in the laboratory and at the computer. Means must be devised for carrying out chamber experiments wherein wall and past-history effects can be minimized. Light intensity and spectral distribution must be controlled. Humidity must be controlled and a wider range of hydrocarbon structures must be examined. Finally field-type experiments must be devised to better follow the loss of SO₂ and the formation of sulfates in the atmosphere.

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Application of Aerometric Measurements and Environmental
Chamber Results to Air Pollution Control Strategies

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Ideally a series of air quality simulation models now should be available to provide the quantitative predictions required to implement air standards. A small group of models are available particularly to treat unreactive pollutants. Even these models require verification for short time prediction periods or for the treatment of extended sources. In general numerous technical problems or experimental data gaps existing including the following: (1) emission source strengths over short time intervals (2) effects of rough terrain (3) wind fields (4) chemical kinetic modules (5) long-range transport (6) removal mechanisms for pollutants (7) comparability between predicted air quality and measured values. At the same time we struggle to solve these problems, prediction requirements expand. For example, the impact of site selections of emission sources varying from power plants and petroleum refineries to shopping centers require evaluation.

Another alternative is the development of empirical relationships based on analysis of data from aerometric networks or laboratory simulations in environmental chambers. These approaches have been used in the past to aid in decision making. Until air quality simulation models are more completely developed, such approaches should continue to be developed and utilized. In the context of this discussion all treatments utilizing "blackboxes" to represent emissions, meteorology or chemical kinetics are considered as empirical relationships rather than air quality simulation models. The most widely used empirical relationship used is that of proportional rollback. The proportional rollback approach is attractive because of its simplicity. However, it is unlikely that this relationship can be justified except as a crude approximation in control strategies for a number of criteria pollutants.

Therefore, it is desirable to discuss several examples of other types of empirical relationships that have been developed.

The upper limit relationship was developed from aerometric measurements to relate oxidant air quality to hydrocarbon emissions. Recently additional evaluation of this relationship has been undertaken for Los Angeles, Chicago, Cincinnati, Denver, Philadelphia, St. Louis and Washington, D. C. The advantages and limitations of this particular relationship will be considered.

A somewhat similar empirical relationship based on another set of aerometric measurements is the sulfate - sulfur dioxide relationship obtained from 24-hr. average monitoring results in U. S. communities. This type of relationship pointed up the difficulties in reducing sulfates to desirable concentration levels even with present sulfur dioxide levels. It allows estimates of the degree of sulfur dioxide control actually required if a sulfate air quality standard should be promulgated

Yet another approach is the use of environmental or "smog chamber" experiments to simulate and predict air quality relationships involving oxidants-hydrocarbons-nitrogen oxides - sulfur dioxide atmospheric reaction systems. Such studies are extremely useful for investigating possible future variations in pollutants which are likely to occur as various control strategies are implemented. Smog chamber experiments are essential to the verification of the chemical kinetic modules of air quality simulation models. Such experiments are anticipated as being very useful in developing models for atmospheric aerosol formation and decay.

Smog chamber results have been combined with estimates of future air quality to predict quantitatively the relative effectiveness of combinations of hydrocarbon and nitrogen oxide control. The limitations on the use of such laboratory simulations will be discussed.

It is important to realize that no one single approach at the present time will necessarily provide the estimations of future air quality needed in relation to control measures. Various empirical relationships must and will continue to be used. The discussion of the limitations of each of these approaches is useful provided that additional constructive insights are obtained. Improvements are most likely to be by small incremental steps. The enormous complexities of the atmospheric systems which we seek to understand cannot but continue to frustrate our continuing efforts to predict the consequences of our actions.

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Abstracts for Davis Conference

EFFECTS OF AIR POLLUTION ON BIOLOGICAL SYSTEMS: AGRICULTURAL CROPS*

by

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Introduction

Air pollution damage to California's agricultural crops has been with us for at least 34 years. Damage was first recognized in 1942 in the San Gabriel Valley when a nurseryman noticed glazing on the under surface of the leaves of petunia plants. Since that time, damage has been observed in many areas of the State and on an increasing number of crops.

There are many types of gaseous pollutants which can cause damage to crops. The easiest classification of such pollutants is one which relates the pollution with the cause; this stated as stationary source and mobile source pollutants. Stationary sources originate as the name implies - from stationary objects such as large industrial complexes. There are two pollutants from this classification which cause plant damage. These are SO₂ (sulfur dioxide) and F (fluorides). Mobile sources originate from the internal combustion engine and related fuels. Ozone (O₃) and PANs (peroxyacyl nitrates) are products of atmospheric photochemical reactions originating from these sources and as such do most of the damage to agricultural crops.

A 1970 statewide survey to determine air pollution damage to agricultural crops indicated that 50% of the damage was caused by ozone, 18% from PAN, and the remainder from other toxicants.

Generally, the various air pollutants cause specific types of symptoms on plant varieties. Although these symptoms can sometimes be confused with symptoms caused from chemical, environmental or other plant stresses, training of the observers can help them differentiate the various problems.

The survey and assessment of air pollution damage to agricultural crops is a difficult task. Essential to any survey of a large area necessitates monetary considerations, manpower needs, and training of personnel. Fortunately, under California conditions we do have a large manpower reserve with county agricultural specialists associated with the County Agricultural Commissioners' staffs and the University of California Extension Service. The Agricultural Commissioners' staffs are presently cooperating in reporting air pollution plant damage in their counties. This information plus field observations by the Department of Food and Agriculture's plant pathologist constitutes the basis for an annual report of crop losses from air pollution. Air pollution losses to agricultural crops in 1970 were reported as \$25,690,679. Figures for 1971 are approximately \$20,500,000; the figures for the remaining years are presently being computed.

Current State of the Art

Losses to California agriculture caused by air pollution injury are currently being computed from visual observations of plant damage and from information of hidden damage on citrus and grapes. Hidden damage or the suppression of

plant growth represents the area of greatest monetary loss to agriculture. The work reported by various members of the staff at the Statewide Air Pollution Research Center at Riverside clearly points this out. In order to have a more accurate assessment of agricultural losses caused from air pollutants, further research is needed concerning the suppression of plant growth and will undoubtedly indicate far greater losses than now reported.

The Department of Food and Agriculture is currently conducting a methodology study to determine the effects of the suppression of plant growth from air pollution. This program is being sponsored by the California Air Resources Board in cooperation with the Statewide Air Pollution Research Center at Riverside.

Research Priorities

1. A model or technique of assessing air pollution damage to plants
 - A. Visual damage
 - B. Suppression of growth;
2. Specific effects on crops - in order of the monetary value in the State's economy;
3. A cooperative study concerning how to fund, report, and utilize information developed
 - A. Land planning use
 - B. Sociological implications
 - C. Health effects, etc.;
4. To develop resistance in various plant varieties to air pollution damage.

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*Abstract of remarks presented at the UC-ARB conference on "Photochemical Smog-- Effects and Causes in Agriculture: Current Status and Priorities in Research," May 20-21, 1974, University of California, Davis.

SUGGESTED QUESTIONS

1. How effective are County Agricultural Commissioner staffs in observing and reporting air pollution damage to crops?
2. How involved is the "UC" in applied research programs in air pollution damage to crops?
3. Is there a standard system for evaluating acute and chronic symptoms?
4. Do cultural practices affect plant symptoms caused by air pollution?
5. Are various California state agencies cooperating in air pollution work and if so, who are they?

EFFECTS OF AIR POLLUTION ON BIOLOGICAL SYSTEMS:

BIOLOGICAL AND PHYSIOLOGICAL EFFECTS*

by

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Major emphasis will be placed on photochemically produced air pollutants, ozone, and peroxyacyl nitrates. Both of these pollutants have serious effects on agriculturally important crops and on vegetation in recreational areas. Little concern has been expressed concerning effects on agricultural animals.

Animals. Commonly observed effects of photochemical air pollutants on humans are eye irritation and a reflex choking response in the upper respiratory tract. In experimental animals, exposure to low concentrations of ozone increases the susceptibility to bacterial infection. This has been attributed to damage to the phagocytic cells in the alveolar sacs and destruction of the enzymes responsible for the digestion of bacteria. An alternative explanation is that one of the two types of cells lining the alveolar sac is particularly susceptible to ozone damage, and thus ozone changes the cell population of the alveolar epithelium. At the biochemical level, two results of ozone exposure have been studied: i) oxidation of unsaturated lipids in the cellular membranes; ii) oxidation of particularly susceptible amino acid residues. The significance of lipid oxidation is indicated by increase in enzyme levels necessary to counteract the effects of the formation of lipid oxidation products. The

*Abstract of remarks presented at the UC-ARB conference on "Photochemical Smog-- Effects and Causes in Agriculture: Current Status and Priorities in Research," May 20-21, 1974, University of California, Davis.

significance of amino acid oxidation is indicated by direct measurement of decreases in lung tissue and by direct measurement of decrease in specific proteins.

As far as research priorities and research programs are concerned, the first requirement would be an assessment of effects on productivity and quality of agricultural animals. Since the producers have not complained about such effects of photochemical air pollution, it is dubious whether a research effort is called for.

Plants. In contrast to agricultural livestock, there is no need for research to detect economic loss to agricultural plants. Current knowledge shows that some crops are more susceptible than others and that in a given crop there may be a range of susceptibility, depending on the variety. Protection of crops by application of chemicals has been a moderately active field of investigation.

Basic research on the mechanism of damage has concentrated on effects of the photochemical air pollutants on the lipids of cellular membranes and on the proteins. Results have been comparable to those described for animals. Is it important to know the chemical mechanism of toxicity of ozone and peroxyacyl nitrates to plants? In a practical sense, only if it helps to develop a cheap antidote or cure. The research priorities would be headed by a need to produce crops without fear of air pollution damage. This goal may be achieved by i) a suitable breeding program to develop resistant varieties; ii) a screening program to produce effective chemical control.

Suggested reading (Books)

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Effects of Photochemical Smog on Ecosystems*

by

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Ecosystems are distinct associations of plants and animals both macroscopic and microscopic together with the physical environment which controls them. The complexity of terrestrial ecosystems ranges from the relatively simple agri-ecosystem comprised of only a few species to the complex natural forest ecosystem with numerous species. In any ecosystem the populations of organisms belong to one of three trophic levels, namely, producers, consumers or decomposers which together form a highly integrated "machine-like" organization for the capture, storage, utilization, and transfer of energy and raw materials. The physical environment provides the energy (solar radiation), raw materials (water and minerals), and living space required to initiate and sustain the growth and reproduction of organisms comprising the ecosystem. The criterion for defining the size or the boundaries of ecosystems is whether the physical and biological parts form an enmeshed unit in which there are defined paths for energy flow and cycling of materials. The good health and productivity of green vegetation at the "producer level" is the key factor which maintains normal function and structure in ecosystem.

The influence of sulfur dioxide and hydrogen fluoride on forest ecosystems has been described; the ongoing examination of the effects of photochemical oxidants on the coniferous forest ecosystem in southern California suggests

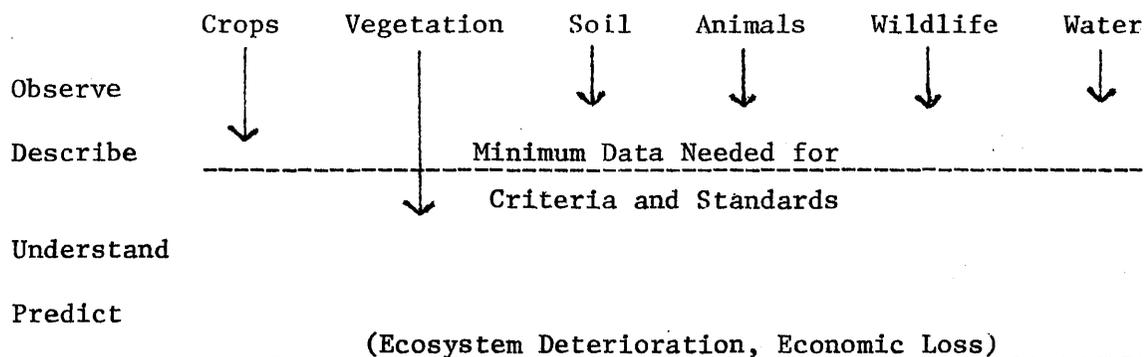
* Abstract of remarks presented at the UC-ARB conference on "Photochemical Smog-- Effects and Causes in Agriculture: Current Status and Priorities in Research," May 20-21, 1974, University of California, Davis.

some similarities. Some of these effects on green vegetation may be summarized: (1) elimination of sensitive species and reduction of diversity in numbers of species, (2) selective removal of larger over story plants and a favoring of plants small in stature, namely shrubs and herbs, (3) reduction of the standing crop of organic matter leading to a reduction of nutrient elements held within the system thus impoverishing the site, and (4) enhancement activity of insect pests and some diseases which hasten tree mortality. Some additional effects on herbivore, carnivore and decomposer populations result when there is a reduction of energy available from green plant production. Herbivores utilize 10-20 percent of the total energy fixed by green vegetation, often in the form of seeds. Seed production is usually drastically reduced after chronic pollutant exposure.

The hydrological balance of a mixed conifer forest may be altered because shade producing evergreen conifers are selectively removed over deciduous species thus, hastening snow melt and increasing evaporative losses. Changes in run off patterns, increased sedimentation and water temperature undoubtedly affect stream fisheries.

Because there are such a multitude of actual and potential pollutant effects to be evaluated in chronically exposed ecosystems it is useful to categorize the steps to be taken in any investigation. In Table I, the logical steps are indicated with emphasis on the fact that the first goal to be achieved is the minimum data needed for establishing reliable criteria for pollutant injury from which suitable air quality standards can be established and defended. The length of the arrows indicates the current relative progress in obtaining the required information:

Table 1. Requirements for evaluating oxidant air pollutant effects on ecosystems.



The ultimate goals of understanding and predicting pollutant effects on ecosystems and evaluating losses calls for a highly integrated systems approach which should be introduced at the earliest possible phase in such an investigation.

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Effects of Air Pollution on Biological Systems:
Ornamentals*

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Priorities for research on air pollution as to its effects and causes, can be significant not only in ornamental plant production, but have much wider implications. Nursery and flower production often occurs in or near centers of air-pollutant generation so damage may be greater and more frequent. The wide range of species and varieties include many sensitive as well as many tolerant to smog. In addition, plants are now considered of increasing importance to our urban and rural landscapes. Plants in cities often are subjected to high levels and/or long periods of pollution. These same plants are looked on as providing respite from the smog that threaten their very existence.

This topic, on a broader scale, was presented in published form last year (1). Two of the authors are this conference's chairman and our keynote speaker.

About all that might be added is to repeat, in modified form, some research areas that appear to be of most importance to ornamental plant production and sales and to the management of landscaped areas. It is acknowledged that efforts need to and are being made to reduce the level of air pollutants. However, their reduction to non-toxic levels at all times for all plants is highly unlikely.

Due to the greater diversity of plants and their high production and maintenance costs on an area basis, research and implementation of research findings will be more feasible for ornamental plants than for more traditional agricultural crops.

Variation in tolerance to pollutants by different seedlings within a species offers considerable opportunity for selecting more hardy plants. Both field and laboratory screening needs to be intensified.

*Abstract of remarks presented at the conference on "Photochemical Smog -- Effects and Causes in Agriculture: Current Status and Priorities in Research", May 20, 21, 1974, University of California, Davis.

Cultural practices that may aggravate or alleviate smog damage need further study. Certain compounds, particularly growth retardants, have increased plant hardiness to air pollutants.

Volatilization of systemically administered chemicals from house and/or landscape plants could be a potential source of pollution.

Pollutants in urban air, particularly the downtown areas, may cause damage to sensitive plants particularly in florist's shops. Air filtration systems might be worthwhile to reduce pollutants to a safe level within retail and wholesale outlets.

New energy sources should be checked closely to be certain pollution is kept to a minimum.

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Agricultural Effects on Air Quality: Solid Waste*

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ABSTRACT

The management and disposal of solid waste from agricultural production and logging debris have an affect on air quality. Information is presented to assess the magnitude and nature of the effect in relation to the various types of sources and management and disposal practices. Thus, problems are identified and their order of importance is suggested in relation to the benefit, cost and time to obtain and implement solutions. The final problem indentification, assessment of research needs and implementation of solutions, and setting priorities are the objectives of the discussion to follow the formal presentation. We all should wish each other "good luck" to achieve the objectives with some degree of satisfaction.

Problem Identification

- A. Solid wastes from agricultural production.
 1. What are they and how might they be categorized?
 2. What is the geographical location and how much is there of each type?
 3. What will they be in the future?
 4. What are current management practices and disposal methods that affect air quality and the relationships of these to effective crop production?
 5. What is the nature of the effect on air quality and the properties of the burden injected into and resident in the air?
- B. Residues from logging, stand improvement and reforestation.
 1. What is the geographical location and amount?
 2. What will they be in the future?
 3. What are current management practices and disposal methods that affect air quality and the relationships of these to effective forest resource and recreation management?
 4. What is the nature of the affect on air quality and the properties of the burden injected into and resident in the air?

The Problems of Solid Waste Management and Disposal that Affect Air Quality

- A. What are they for agricultural production?
- B. What are they in the forest?

*Abstract of remarks presented at the UC-ARB conference on "Photochemical Smog-- Effects and Causes in Agriculture: Current Status and Priorities in Research," May 20-21, 1974, University of California, Davis.

Assessment of Research Needs

- A. What is the meteorological situation with regard to A.5. and B.4. above?
- B. Are there likely to be new uses of the wastes implemented that will reduce or eliminate the burden injected into the air?
- C. What information is needed to solve the problems? What will it cost to acquire it and implement the solution?
- D. What is the nature of the value of the benefit? Its cost? How should it be paid for?

Priorities

- A. What are they for the identified problems?

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EFFECTS OF PESTICIDES ON AIR QUALITY (AND VICE VERSA).^{*} Donald G. Crosby,
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While the presence and movement of pesticides in air as "drift" have been recognized and investigated for decades, it is only during the past several years that the photodecomposition of pesticides under simulated atmospheric conditions has been demonstrated; actual observation in the atmosphere was achieved only last October. In no instance has the environmental fate of any of the 900 registered pesticide chemicals been accounted for, and knowledge of their atmospheric movement and transformations often could influence importantly the application of existing chemicals and the introduction of new ones.

Our current discoveries raise the following priority questions and suggest the indicated research needs (among others):

- I. Basic Issues.
 - A. What fundamental (photochemical) reactions do pesticide vapors undergo in the sunlit atmosphere? [Improved atmospheric modelling; laboratory identification of photoproducts; rate measurements].
 - B. What part do airborne particulates play in pesticide transformations? [Chemical composition and structure of particulates; photoreactions on surfaces; atmospheric simulation; source and movement of contaminated dust].
 - C. How can pesticides and their transformation products be identified and measured in the atmosphere? [High-volume air-sampling; submicro identification methods; specific analytical detectors].
- II. Extensions
 - A. What order of research priority should any particular pesticide receive?
 - B. What are the "terminal residues" in each case, and what happens to them?
 - C. What other variables are involved? [Interactions with photochemical oxidants; effects of climatic factors--temperature, humidity, rainfall; relative importance of sources--drift, volatility, dust, smoke].
- III. Applications
 - A. What is the toxicological significance of the pesticides and photoproducts--in air and precipitated?
 - B. How will this influence registration and application?
 - C. What place do atmospheric transformations have in the world-wide fate of pesticides?
 - D. Could atmospheric photodecomposition be used to destroy unwanted pesticides?

^{*}Abstract of remarks presented at the UC-ARB conference on "Photochemical Smog--Effects and Causes in Agriculture: Current Status and Priorities in Research," May 20-21, 1974, University of California, Davis.

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D.G. Crosby
May, 1974

AGRICULTURAL EFFECTS ON AIR QUALITY: ANIMAL WASTES^{1/}R. E. Luebs^{2/}

ABSTRACT

Gases emanating from animal waste have been of increasing concern to researchers and the public during the past five years. This results from the increasing size and concentration of domestic animal enterprises, changes in waste management practices, the close proximity of some of these operations to urban residential areas, and an increasing concern for quality of the environment. Odor is currently the major air pollution problem caused by animal wastes. Odor from animal waste usually is a composite of odors from several gases.

Significant aspects of odor problems with animal waste are as follows:

1. Measurement of animal waste odor.
2. Odor intensities that will be tolerated by people not involved in animal industry.
3. Odor intensities that can be expected from different management practices.
4. Identification of malodorous gases.
5. Measurement of atmospheric concentrations of malodorous gases.
6. Volatilization rates of malodorous gases from animal waste areas.
7. Meteorological effects on the distribution of malodorous gases in the atmosphere.
8. Identification and effect of factors involved in formation of malodorous compounds.
9. Odor control.

Many instances of legal action against animal producers because of odor problems have been recorded. Standards for permissible odor intensity in the vicinity of animal operations have not been defined. Initially, it will be necessary to set standards based on olfactory measurement because (1) some odors are detectable by this method at gas concentrations which are too low for current physico-chemical techniques and (2) research has not

^{1/} For presentation and discussion at the University of California--California Air Resources Board Conference, Davis, Calif., May 21, 1974.

^{2/} Soil Scientist, Agricultural Research Service, USDA, and Associate in Experiment Station, University of California, Riverside.

yet correlated intensity of odor with known concentrations of malodorous gases in a mixture.

The following compounds have been identified as volatilizing from animal wastes: carbon dioxide, methane, ammonia, hydrogen sulfide, aliphatic amines, fatty acids, organic sulfides, mercaptans, carbonyls aldehydes, ketones, indole, and skatole. Most, but not all of these gases are odorous. All odorous gases may not yet have been identified. Atmospheric concentrations in the vicinity of animal waste have been measured for only a few of the known compounds. Most of the identified gases have been reported from studies with poultry and swine waste.

Odor control currently involves changing waste management practices and the possible use of chemicals as oxidants, deodorants, or masking agents. Total elimination of odor in animal waste operations is unlikely.

Several additional significant aspects of the relationship between animal waste and air pollution are as follows:

1. Dust from the surface of cattle feedlots and corrals.
2. Absorption of gases volatilized from animal waste by surface waters.
3. Effects of gases volatilizing from animal waste on animal health.
4. Interaction of gases emanating from animal waste and from industrial or other sources.

Atmospheric dust from beef cattle feedlots has been a problem in dry areas. Dust can increase the incidence of animal respiratory disease. It may also increase the odor problem. Dust can be reduced by sprinkling, windbreaks, or increased density of animals. Ammonia, which volatilizes from animal waste in large quantities, is readily absorbed by surface waters. If nitrogen is limiting for algal growth in surface waters, high atmospheric concentrations of ammonia emanating from animal waste could lead to eutrophication. Ammonia flux or volatilization rate from animal wastes in lots or corrals has not been measured. High concentrations of ammonia, and possibly hydrogen sulfide in poorly ventilated animal housing, have been reported to be deleterious to animal health. Little is known about the atmospheric interaction of chemical species emanating from animal wastes and from other sources. Sulfuric acid and ammonium sulfate aerosols are likely to be present where large areas of animal waste exist.

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APPENDIX C

UC-ARB Conference reports appearing in Statewide
Coordination Newsletter April, 1974 and May, 1975

STATEWIDE COORDINATION NEWSLETTER



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STATEWIDE AIR POLLUTION RESEARCH CENTER

University of California: Berkeley • Davis • Irvine • Los Angeles • Riverside • San Diego • San Francisco • Santa Barbara • Santa Cruz

April 1974
Issue No. 2

UC-ARB BERKELEY CONFERENCE

The first of a series of conferences sponsored jointly by the California Air Resources Board and the University of California, through the SAPRC and Statewide Coordination for Air Pollution Research was held in Berkeley, California, March 18-19, 1974, entitled "Chemical and Physical Interactions of SO_x, NO_x, and Photochemical Oxidant: Current Status and Priorities in Research." It was chaired by James N. Pitts, Jr., Director, SAPRC and Statewide Coordination. Attending the two-day conference were approximately 70 experts concerned with this area of research, coming from organizations such as the University of California, the California Air Resources Board, Environmental Protection Agency, other control agencies, and industries.

The first morning's program began with an overview by John F. Finklea, M.D., on "Health Effects and Atmospheric Transformations in the SO_x, NO_x, and Photochemical Oxidant System." SO_x and NO_x from stationary and mobile sources were then discussed by other speakers. The afternoon session centered on physical and chemical transformations in aerosol systems, the kinetics and mechanisms of SO_x, NO_x, hydrocarbon and oxidant systems, and their implications in modeling photochemical smog.

The second morning's program emphasized discussion of analytical techniques, the validity of aerometric data in ambient atmospheres, and chamber studies in the NO_x and hydrocarbon and oxidant systems. Dr. Paul Altshuller, of the EPA, closed Tuesday afternoon with a discussion of chamber studies in the SO_x, NO_x, and hydrocarbon system, and the application of aerometric and chamber data in control strategies.

The participants felt that the technical discussions were highly beneficial in delineating research priorities and technical problem areas. A short written report on the contents of the conference will be produced by the Statewide Coordination Office at the SAPRC.

FUTURE UC-ARB CONFERENCES

Planning is completed for the upcoming conference on the interrelationships of photochemical smog and agriculture, to be held May 20-21, 1974, Mrak Hall, UC Davis campus. Its chairman will be Dr. O. C. Taylor, Associate

Director of SAPRC, and the title is "Photochemical Smog--Effects and Causes in Agriculture: Current Status and Priorities in Research." The keynote speaker for the Davis conference is Dr. Walter Heck, Agricultural Research Service, USDA, who will speak on "Priorities for Future Biological Research in Air Pollution." The Davis conference will consider two aspects of the interrelationship of agriculture and air pollution: (1) the effects of air pollution on biological systems, with respect to field crops, biochemical and physiological systems, the ecosystem, and ornamentals; and (2) the effects of agricultural practices on air quality, through solid waste, pesticides, herbicides, and animal waste.

As a result of the success of the technical conference in Berkeley, ARB and SAPRC personnel agreed that a substitution should be made for the large general conference previously planned for September 23-24, 1974, at UC Riverside. Instead of a "maxi" conference, a small technical conference will be conducted on the UCR campus in December 1974, entitled "Oxidant Control by Reductions of Hydrocarbons and NO_x." We think this revision will be more rewarding, both scientifically^x and technically, to the California Air Resources Board as well as to the University of California, control agencies, and the industries involved.

The final conference in this series will be held in January 1975 on "Health Effects Associated with Air Pollution," under the chairmanship of T. Timothy Crocker, M. D., on the UC Irvine campus.

Anyone who is interested in attending any of the conferences in invited to contact Dr. William C. Kuby, Associate Director, Statewide Coordination for Air Pollution Research, SAPRC, University of California, Riverside, CA. 92502, phone (714) 787-5128.

GRANTS AND CONTRACTS FOR AIR POLLUTION RESEARCH IN THE UC SYSTEM

In the previous issue of the Statewide Coordination Newsletter (Issue No. 1, February, 1974) we included a list of grants and contracts on air pollution research being conducted by faculty and staff on the various campuses of the University of California. We invited additions to that listing of any current UC research on air pollution that was not cited. We have received the following information of research grants to be added to the previous listing. We invite others to notify us of any additional grants or contracts which might have been missed.

STATEWIDE COORDINATION NEWSLETTER



STATEWIDE AIR POLLUTION RESEARCH CENTER

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May 1975
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AIR POLLUTION RESEARCH NEWS: HELP WANTED!



We welcome contributions to the Statewide Coordination Newsletter of news items which might be of interest to the University of California air pollution research community. Please send information to Dr. James N. Pitts, Jr., Director, SAPRC, University of California, Riverside, CA 92502. In addition, any ideas for improving intercampus communication on air-pollution-related matters would be most welcome.

UC-ARB RIVERSIDE CONFERENCE

The third of a series of conferences during 1974-75, sponsored jointly by the California Air Resources Board and the University of California--through the Statewide Air Pollution Research Center and Statewide Coordination for Air Pollution Research--was held December 16-17, 1974, UC Riverside, in the Large Dining Room of the Commons. The conference title was "Technical Bases for Control Strategies of Photochemical Oxidant: Current Status and Priorities in Research," and it was chaired by Dr. James N. Pitts, Jr., Director of SAPRC.

The Monday morning session was devoted to "Medical Bases for Control Strategies." The keynote speaker for the Conference, Dr. David V. Bates, Dean of the Faculty of Medicine, University of British Columbia, Vancouver, reviewed the contemporary knowledge concerning the effects of ozone on animals and man and particularly emphasized the difficulty of predicting whether or not contemporary knowledge concerning the effects of ozone on animals and man and particularly emphasized the difficulty of predicting whether or not contemporary levels might or might not produce long-term effects on the lung. It is possible that humans may adapt to some aspects of toxicity of ozone and that their reactions to ozone might be conditioned to whether or not they have lived in a high ozone environment. Healthy students from Canada showed more symptoms, such as substernal pain and short cough, than did similar individuals who lived in Long Beach, California. Occurrence of sulfur dioxide with ozone caused a marked exacerbation of symptoms, showing that both pollutants together caused much greater effects than their sum separately.

Dr. Bertram Carnow, Director, Occupational and Environmental Medicine, University of Illinois, School of Public Health, cited 35% "excess" deaths in Chicago on days when smog was heavy, implicating a combination of ozone plus sulfates. He called for expanded work on this combination to find out how much injury can be expected with these two pollutants, and believes there is a need for an air quality standard for H_2SO_4 and acid sulfate. Dr. Carnow reviewed the growing body of knowledge about the cumulative effects of different pollutants. For example, sulfur dioxide and ozone (in combination) have been shown to cause pulmonary insufficiency, and particulates (in combination with nitrogen dioxide) have caused inflammation of the bronchioles in animals.

The final speaker at the Monday morning session, Dr. John H. Knelson, Director, Human Studies Laboratory, U. S. Environmental Protection Agency (EPA), believes that the present Federal Air Quality Standard of 0.08 ppm oxidant is not too stringent and should be maintained. The EPA has in progress a comprehensive research program designed to provide further information on this question, including clinical studies designed to define the influence of ozone on (a) cardiopulmonary function in healthy and susceptible individuals with and without thermal stress and exercise, (b) psychophysiological functions, (c) cytogenetic status, (d) status of immune mechanisms, (e) carcinoma-associated antigen titers, and (f) serum enzyme levels reflecting organ damage.

The current EPA program is confirming results upon which the current oxidant standard is based. In addition, results not previously described in humans are being found, such as immunological and cytological variations.

The first Monday afternoon talk was given by Dr. James N. Pitts, Jr., who focused on recent experimental data obtained from the SAPRC smog chamber facility. For example, the addition of formaldehyde to a butane/ NO photolysis accelerated in the rate of conversion of NO to NO_2 and the buildup of ozone. The sensitivity of a computer kinetic model to the value of the rate constant for the reaction $OH + NO + M \rightarrow HONO + M$ was illustrated. The recent results obtained at SAPRC for this rate constant improved the fit for the first two hours of the NO_2 production curve.

Spectra obtained by combining a high resolution Fourier infrared interferometer with the in-situ longpath optical system in the SAPRC evacuable chamber were shown. The photolysis of a mixture of NO , NO_2 and propylene in air showed infrared bands assigned to PAN, ethyl nitrate, formic acid, nitrous acid and nitric acid. Additionally, several ozone-olefin reactions in the dark produced spectra of a variety of species including ketene, and possibly peroxyformic acid and α -carbonyl hydroperoxides.

Finally, the recent oxidant measurement calibration controversy was discussed. After briefly outlining the history of this problem, Dr. Pitts reported recent work at SAPRC in which the ARB potassium iodide calibration method was investigated using infrared absorption

by ozone as a standard. In order to apply the only absolute infrared absorptivity data for ozone available in the literature, a study of the dependence of the absorptivity on spectral resolution was carried out. Using the absorptivity determined for the comparatively high resolution afforded by modern infrared instruments, the SAPRC results for the ARB method fell in between those of the EPA and the ARB ad-hoc committee, namely that for ozone in dry air the ARB method was approximately 10% high while for wet air (~50% RH) it was approximately 20% high. Dr. Pitts emphasized the need for further studies to determine to everyone's satisfaction which method (ARB, LAAPCD, or EPA) actually gives the most accurate and reproducible values in the ambient concentration range. However, he pointed out that regardless of which is the best measurement technique, previous conclusions that ambient oxidant levels are highest in the eastern end of the South Coast Air Basin, are shown to be incorrect when all of the oxidant air monitoring data from the Basin through the year 1973 are placed upon a common calibration basis.

Dr. Basil Dimitriades, Chief of the Atmospheric Reactivity Section of the EPA, suggested that smog chamber data constitute the only source of direct evidence regarding the dependence of photochemical oxidant on the oxidant precursors; atmospheric data are inadequate in showing this dependence. He supported continuing investigation along two lines-- very close simulation of the real atmosphere in the chamber (the "close simulation" approach), and studying the degree to which data taken in a certain chamber system reflect real atmospheric phenomena (the "direct" approach). A third approach which has been used by Los Angeles County-- empirical correlation of chamber data with aerometric data, is judged unacceptable, according to Dimitriades.

Both the "close simulation" and "direct" approaches should be pursued in parallel, in view of the indispensability of the smog chamber methodology and data and the good prospects for successful application to control strategies. Dr. Dimitriades particularly called for further tests to determine the effect of chamber operating conditions on the ozone-NO_x-hydrocarbon relationship and further field tests in urban and nonurban atmospheres, both to guide the design of chamber experiments and to compare the chamber and atmospheric data.

The final talk of the afternoon on the use of ambient air data to develop control strategies was presented by Dr. A. Paul Altshuller, Director of the Chemistry and Physics Laboratory of the EPA. Dr. Altshuller felt that use of ambient air data is a useful approach in the development of oxidant control strategies, although a significant limitation in its use has been inadequacies in the ambient air data base. Low quality measurements, lack of data for some significant pollutants or for necessary meteorological parameters, or insufficient time span have limited the utility of the data.

Dr. Altshuller reported a preliminary investigation of the inter-relationship of aerometric data, modeling computations, and smog chamber experiments using oxidant-NO_x-hydrocarbon data from Philadelphia,

Washington, D.C., and Denver; computer modeling data from Dr. Marcia Dodge, EPA; and smog chamber data from Dr. Dimitriades. The results, although preliminary, showed reasonable agreement for several hydrocarbon-NO_x reactant combinations.

Robert Neligan, Director, Office of Air Quality Planning and Standards, EPA, began the Tuesday morning session with a discussion of "The Validity of the Strategy of Linear Rollbacks of Hydrocarbons to Achieve Oxidant Air Quality Standards," pointing out the well-known limitations of this strategy.

Upper limit curves relate daily oxidant maxima to the levels of morning hydrocarbon emissions. When developed for a specific city or air basin, their form reflects the effects of meteorology, chemistry, and transport as crude averages. If the curve is based on data taken at many air monitoring stations located within an air basin, the resulting relationship somewhat reflects the results of atmospheric transport within that basin.

Recent EPA studies in Ohio have shown that nonurban oxidant levels are much higher than can reasonably be explained by local levels of hydrocarbon emissions. Analysis of oxidant data obtained using airplanes and a system of ground-based stations clearly shows that local oxidant levels can be explained only if oxidant transport is occurring over large distances from places as remote as the metropolitan regions of the eastern U. S. seacoast. These data suggest that meeting the Federal Air Quality Standard for oxidant in southern Ohio may be impossible if control is based solely on linear rollback of local hydrocarbon emissions.

In his talk, Dr. Robert F. Sawyer, Professor of Mechanical Engineering, University of California, Berkeley, pointed out that catalyst-equipped 1975 cars initiate a substantial reduction in automotive hydrocarbons and CO and offer somewhat improved performance and fuel economy, compared to 1974 models. Substantial increases in fuel economy are most easily obtained, not by relaxation of emission controls, but by reductions in vehicle weight.

Sawyer discussed several research areas, including particle production from the catalyst, conversion of bound fuel sulfur to sulfate particulate, formation of aldehydes and odorants, improved control of evaporative emissions, and re-evaluation of the NO_x emission standard, with possible relaxation to 1.0 to 1.5 gm/mile to encourage the development of stratified charge engines.

The final speaker Tuesday morning was Dr. Henry K. Newhall, Professor of Mechanical Engineering, University of Wisconsin, Madison, who said that, in the next decade, stratified charge engines appear to offer the most promising alternative to the present I.C. engine. Stratified charge engines have been developed by Honda, Ford, and Texaco and are under study by most large automobile manufacturers.

Tests of prototype engines show that stratified charge engines can meet the 1977-78 hydrocarbon and CO emission standards with good fuel economy. They can also meet the 0.4 gm/mile NO_x standard by sacrificing fuel economy. The decrease in fuel economy increases as the NO_x standard is decreased below 1.0 gm/mile.

On Tuesday afternoon, December 17, five speakers discussed the topic "Emissions Inventories, Transport, and Models in Control Strategy Formulations."

Dr. Donald Blumenthal, of Meteorology Research Inc., reviewed recent surveys from aircraft designed to clarify the role of vertical and horizontal transport and mixing in accounting for observations on oxidant and other pollutants. He found that the vertical distribution of pollutants is highly complicated. Layers aloft containing ozone often persist overnight, only to be remixed with the surface air on the succeeding day. This can account for the appearance of oxidant at a very early hour in the morning when there is ozone aloft but in the stable ground layer of air, nitric oxide from auto exhaust has destroyed the ozone during the night hours. Blumenthal discussed data for a famous episode on July 25, 1973. One statistic he cited was that approximately 185 metric tons per hour of ozone were being transported eastward across a line between Pomona and Corona. This is enough to account for an average of about 0.25 ppm ozone over the whole eastern South Coast Air Basin.

Mr. Robert Lunche, Air Pollution Control Officer of the County of Los Angeles, emphasized the fact that progress in controlling photochemical smog has been made. . . "eye irritation is less frequent and less severe. . . damage to vegetation has been markedly reduced, and ozone (or oxidant) concentrations have been decreasing in coastal areas since 1964. . . and in inland areas 40-60 miles from the coast since 1970 or 1971." As part of his presentation, Lunche showed a contour diagram of oxidant vs. hydrocarbons and nitrogen oxides. He used it to explain the District's control strategy for oxidant pollution and his prediction that "control of motor vehicle emissions in the proper manner will achieve compliance with the oxidant and all other air quality standards except those for particulates by 1980 to 1985." This strategy emphasizes control of hydrocarbons rather than nitrogen oxides. The District's view is that the standards will be achieved more rapidly if this course is taken, rather than a course which would call for equal emphasis on both pollutants, such as reduction in vehicle miles traveled.

Mr. John Kinoshian, Chief of the Division of Technical Services, California Air Resources Board, presented trends in the measured values of oxidant in the South Coast Air Basin and their implications for control strategies. Discernment of trends is hampered by year-to-year fluctuations due to the variability of the weather. Nevertheless, Kinoshian also believes that oxidant concentrations decrease slightly in inland areas in the South Coast Air Basin during the past four years. This conclusion will not be affected, even after the recent discrepancy between the Los Angeles oxidant measurement procedure and the ARB oxidant

measurement procedure is corrected, since it is presumed that all agencies have used self-consistent procedures. The slight decline in oxidant trend in the inland areas of the South Coast Air Basin may be attributable to weather, as well as to hydrocarbon control or increased emissions of oxides of nitrogen.

The first of the final two afternoon papers on "The Use of Airshed Models in Developing and Testing Control Strategies," was given by Dr. Ralph Sklarew of Xonics, who discussed the development of mathematical models to estimate the impact of various control strategies on air quality. He said, "Airshed models have not been really used in developing and testing control strategies for photochemical oxidant. Lumped parameter chemical mechanisms for photochemical smog have had some success. When carefully tuned for a specific episode for a few hours, predictions of maxima can be obtained within 20% of the measured concentration."

In the last presentation of the conference, Dr. Alan Eschenroeder, of General Research Corporation, (now with Environmental Research and Technology, Inc.) listed several different applications of GRC's DIFKIN photochemical diffusion model of the air quality. His discussion dealt with some of the limitations of modeling, emphasizing the complexities of source distribution, meteorology, and air movement and chemistry which should be included for any complete description of atmospheric photochemistry. He presented data which showed that the photostationary state is not obtained in the real atmosphere, perhaps because of "mixedness effects" on chemical rates.

UC-ARB IRVINE CONFERENCE

The fourth and last of the current series of UC-ARB Conferences, "Health Surveillance Related to Air Pollution" was convened March 25, 1975 under the chairmanship of Professor T. Timothy Crocker, Chairman of Community and Environmental Medicine, UC Irvine. The conference was held in Costa Mesa and followed a meeting of the Air Quality Advisory Committee, California State Department of Health. Dr. John Goldsmith, chairman, and members of the latter committee participated, as well as spokesmen from the areas of meteorology and air monitoring, including particle characterization. The objective was to review the technologies required in developing a health surveillance program applicable in the California South Coast Air Basin.

Particle size and composition data, localized vs. basin-wide accumulations of specific pollutants, pollutant transport patterns and coincidence of gaseous and particulate pollutants in sites throughout the basin were reviewed. It was agreed that monitoring technologies now available could be deployed more generally by existing agencies or by in-state groups on contract. Mr. Walter Crider of EPA conducted a tour of a CHAMP monitoring station on the day following the conference.

Epidemiologic approaches were reviewed, including the CHES program, (a surveillance of lung function in children, exacerbation of disease in heart-lung patients and asthma panels), the hospital-admissions pilot program in Riverside, the breathmobile study of UCLA and the Los Angeles County Lung Association. Methods for evaluation of lung function were reviewed with regard to the level of reliability, sturdiness, ease of performance by age group, and correspondence with clinically recognized disability. No single approach was agreed upon as preferred over all others, but the merits of each type were evaluated. Consensus was strong that the 1971 through 1973 data from the CHES study in California should be analyzed at an accelerated pace. Dr. Gory Love of the National Environmental Research Center, EPA, participated in the conference and expressed readiness to cooperate in pursuing the latter objective.

A follow-on conference can be held to develop specific recommendations for a health surveillance program if an appropriate call is made by an agency prepared to fund such a program.