Oxidant Modeling Status

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Mathematical models that relate pollutant emissions to ambient air quality through the theoretical treatment of the chemical and physical processes of the atmosphere are reviewed. An evaluation of currently available models is presented and various shortcomings of state-of-the-art models are discussed. The program to verify the model for the St. Louis Regional Air Pollution Study is outlined and a discussion of the approach is presented.

Pollutants emitted into the atmosphere are transported, dispersed, transformed, and deposited via complex physical and chemical processes. Mathematical models provide a unique technique for evaluating the impact of anthropogenic emissions on air quality and will prove to be formidable tools in implementing the mandates of the Clean Air Act of 1963 and its amendments. Air quality simulation models (AQSM) provide the most fundamental approach to relating emissions to air quality through mathematical descriptions of the physical and chemical processes operating in the atmosphere.

The chemical mode of an AQSM is either nonreactive or reactive. Models that only consider inert pollutants or pollutants whose chemical transformation can be expressed by a first order reaction rate are nonreactive. Typical nonreactive pollutants are total suspended particulates, sulfur dioxide (SO_2), carbon monoxide (CO), and sulfate ($SO_4^=$). Reactive models, the subject of this review, are termed here as photochemical air quality simulation models (PAQSM). This is because of the major role played by photolytic reactions in the atmospheric transformation processes of pollutant species. Pollutants typically considered in PAQSM include reactive hydrocarbons (HC), CO, nitric oxide (NO), nitrogen dioxide (NO_2), and ozone (O_3).

MODEL FORMULATION

The development of a mathematical relation for simulating the transport, dispersion, transformation, and disposition of pollutant emissions into the atmosphere can assume varied ranges of complexity and scale, depending on the application under consideration. The need for practical PAQSM for describing the physical and chemical dynamics of the atmosphere necessitates the development of simplified approaches for treating reactive species in the turbulent planetary boundary layer. The basis of all PAQSMs is the equation of conservation of mass, which, with certain simplifying assumptions, reduces to the so-called atmospheric diffusion equation (1):

$$(\partial\bar{c}_i/\partial t) + [\bar{u}(\partial\bar{c}_i/\partial x)] + [\bar{v}(\partial\bar{c}_i/\partial y)] + [\bar{w}(\partial\bar{c}_i/\partial z)] = (\partial/\partial x)\{[(K_H\partial\bar{c}_i)/\partial x]\} + (\partial/\partial y)\{[(K_H\partial\bar{c}_i)/\partial y]\} + (\partial/\partial z)\{[(K_z\partial\bar{c}_i)/\partial z]\} + [R_i(\bar{c}_i \ldots \bar{c}_n, T)] + [\bar{S}_i(x, y, z, t)] \quad (1)$$

where

$\bar{c}_i$ = ensemble mean concentration for species i,
x, y, z = Cartesian coordinates,
$\bar{u}$, $\bar{v}$, $\bar{w}$ = ensemble mean velocities,
K_H, K_z = horizontal and vertical eddy diffusivities,
$\bar{S}_i$ = rate of injection (or removal) of species i by a source (or sink),
R_i = rate of production (or consumption) of species i through chemical reactions,
T = temperature, and
t = time.

The set of equations described by Equation 1 can be solved numerically to provide the theoretical mean concentrations of species i as a function of location and time. The model equations require (a) input information for initial and boundary concentrations of each of the pollutant species considered; (b) meteorological data for prescribing the wind field, turbulent diffusivities, solar radiation, and mixing height; and (c) source emissions as a function of location, time, and composition. Spatial scales may range from micro to synoptic and can be classified by the following four functional modeling domains (note that H = mixing height and PBL = height of the planetary boundary layer):

Scale	Domain	Dimension
Local	Micro	200 m x 200 m x 100 m
Urban	Sub-meso	50 km x 50 km x H
Regional	Meso	1000 km x 1000 km x PBL
Continental	Meso-synoptic	3200 km x 2400 km x PBL

Historically, the development of PAQSM has focused on the urban scale, where the O_3 problem was thought to originate and reside. Recent field programs indicate that O_3 and its precursors are not confined to the urban complex but in many instances are transported over hundreds of kilometers and time periods of several days. Based on these findings, development work in regional PAQSMs has begun. Given that the majority of research and development has concerned the urban problem, most of this review and evaluation will focus on that scale. Three basic approaches to photochemical air quality simulation modeling have evolved over the past several years. They are

1. A grid model based on numerical solution of the coupled atmospheric diffusion equations in three spatial dimensions on a grid over the region of interest,
2. A trajectory model based on simulating chemistry and vertical transport in air column advection with the local mean wind velocity, and
3. A box model based on simulating chemical processes in a well-mixed region in which no spatial inhomogeneities are assumed to exist and within which emissions are mixed instantaneously throughout the region.

Status of Urban Oxidant Models

A synopsis of air quality simulation models for photochemical oxidants is given below. The models considered are a representative sample of the current state of the art, limited to urban scales, and deterministic in nature.

1. Three-dimensional grid model based on numerical solution of the atmospheric diffusion equation. Three-dimensional wind field derived from ground-level measurements. Pollutants emitted from ground-level

sources are injected into the bottom layer of grid cells; emissions from stacks are distributed among the grid cells aloft; numerical solution by method of fractional steps with advection treated by SHASTA algorithm; vertical diffusion and chemistry by Crank-Nicholson method. A 35-step chemical reaction mechanism based on the reactions and reactivities of carbon bond types is used. Development of the model has been completed. Estimated completion of the regional air pollution study (RAPS) model verification is November 1978 (2, 3, 4, 5).

2. Two-dimensional grid model based on the numerical solution of the vertically integrated atmospheric diffusion equation; a modified Gear method is used in the numerical integration of coupled ordinary differential equations. The model utilizes a mass consistent wind field approach for generating detailed wind fields, vertical profiles of velocity, and diffusivity. Pollutants emitted at ground level and aloft are injected uniformly into the appropriate well-mixed cell. A 48-step generalized lumped chemical mechanism based on hydrocarbon reactivity is used. Development of the model has been completed. Estimated completion of RAPS model verification is November 1978 (6).

3. Trajectory model based on a moving column of air in which vertical diffusion and chemical reactions take place. Pollutants are emitted into the appropriate vertical cell. The column of air follows a surface trajectory interpolated from surface wind data. A modified Gear method is used in the numerical integration of coupled ordinary differential equations. A 46-step generalized lumped chemical mechanism based on hydrocarbon structural classes is used. Development of the model has been completed. Estimated completion of RAPS model verification is January 1979 (7, 8).

4. Single well-mixed cell. Coupled ordinary differential equations that include emission, advection, entrainment, dilution, and chemical reactions are solved using the Gear method. Emissions, which are considered horizontally homogeneous, mix instantaneously within the temporally prescribed mixed layer. A 36-step generalized lumped chemical mechanism based on hydrocarbon structural classes is used. Development of the model has been completed. Estimated completion of RAPS model verification is June 1978 (9).

Table 1. Sources of error in PAQSMs and estimated impacts on predicted maximum O_3 concentration.

Source of Error	Uncertainty in Predicted Maximum O_3 Concentration (%)
Chemical mechanism	
Reaction rate constants	±20
Structural or reactive hydrocarbon classification	±50
Unknown reactions	±10
Meteorology	
Wind speed and direction	±20
Mixing depth	±25
Light intensity	±20
Initial and boundary condition	
Horizontal boundary	±20
Vertical entrainment	±50
Initialization	±20
Source emissions	
Spatial resolution	±20
Temporal resolution	±10
Emission factors	±30
Hydrocarbon composition	±20

The evaluation of AQSMs has been to assess both the validity of the basic assumptions used in deriving the models' working mathematical equations and potential sources for inaccuracies in the treatment of meteorological, chemical, and emission phenomena by the model (10, 11, 12, 13). A summary of these sources of error and an estimate of their impact on the predicted maximum O_3 concentration of a model is presented in Table 1. Several models have undergone limited verification studies (4, 6, 14, 15), as summarized in Table 2 (13). The reported results from these studies appear promising. Prior to extensive model application studies, further model verification and evaluation is warranted. The verification program under consideration as part of RAPS should provide the extensive model testing that is needed.

Model Verification and RAPS

Model verification studies have been limited in nature due to the lack of adequate air monitoring data bases against which to test models. The unverified status of AQSMs has proved a considerable deterrent in their application, both in terms of the rather extensive data resources required in operating the models and the unknown accuracy of their performance. Uncertainty limits on model prediction inaccuracies caused by all sources of error can be obtained through extensive comparison of model concentration predictions and ambient measurements. Comparisons should be made for a variety of meteorological and, where possible, emission conditions. Exercising models that have a sufficient data base to establish model prediction accuracy and uncertainty limits will provide insight into establishing the degree to which models can be extrapolated to conditions beyond those in the domain of evaluation.

The St. Louis RAPS (16), a 5-year field program sponsored by the U.S. Environmental Protection Agency (EPA) begun in 1972, will provide highly resolved spatial and temporal emissions and ground monitoring data for use in model verification studies. Three major objectives sought by the program are

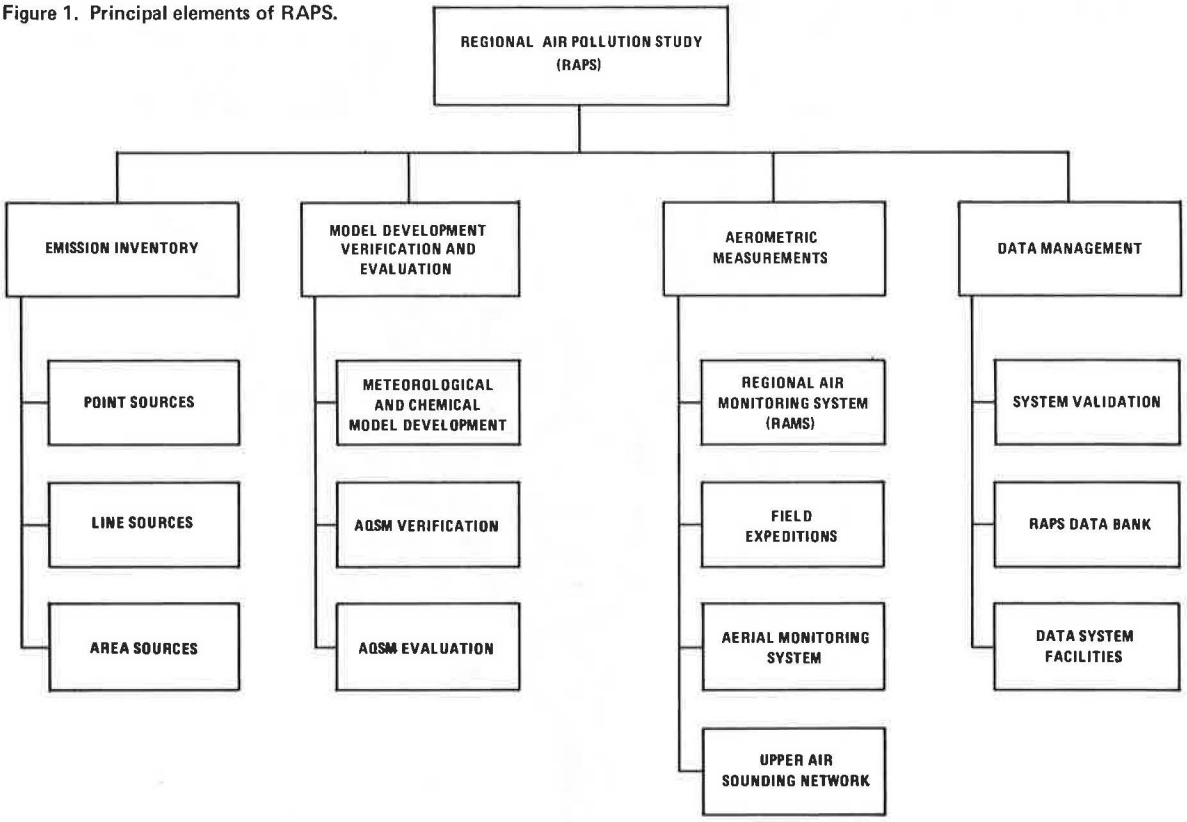
1. To develop, evaluate, and verify air quality simulation models on an urban scale covering urban and rural stationary and mobile sources;
2. To develop, evaluate, and verify models of local phenomena that complement urban models; and
3. To archive all data collected under the program in the form of a readily retrievable data base to use in evaluating future air quality simulation models.

The principal elements of RAPS are shown in Figure 1. The monitoring network that developed late in 1974

Table 2. Previous photochemical PAQSM verification studies.

Region	Time Periods	Pollutants Compared
Denver. Portions of south coast air basin. Both 80 × 80-km (50 × 50-mile) and 129 × 161-km (80 × 100-mile) regions.	6 d in 1969; June 26, 1974	Studies performed with both the 15- and 31-step kinetic mechanisms. In both versions, pollutants compared were NO, NO_2, O_3, CO, reactive and unreactive hydrocarbons.
San Francisco Bay area. 170 × 210-km (106 × 130-mile) region. A variety of subregions and grid sizes (1 to 5 km) were employed.	July 26 and 27, 1973; August 20, 1973; September 26 to 28, 1973	Pollutants compared were NO, NO_2, O_3, CO, reactive and unreactive hydrocarbons.
Trajectories in south coast air basin.	6 d in 1969 LARPP data	Pollutants compared were NO, NO_2, O_3, CO, reactive and unreactive hydrocarbons.

Figure 1. Principal elements of RAPS.



consists of 25 monitoring stations, which are spatially distributed in a spiral configuration covering approximately a 6400-km^2 area. Figure 2 shows the geographical locations of regional air monitoring stations (RAMS) in the St. Louis area, and Table 3 gives the parameters measured and the instrumentation used at each station. Operational schedules for the upper air sounding network are as follows:

Sounding	Normal Operation	Intensive Study Periods
Rawinsonde	5 d/week, 4/d (6-h intervals) to 3.0 km; two locations	7 d/week, 4/d (6-h intervals) to 3.0 km; four locations
Pibal	20/d at hourly intervals between radiosondes; two locations	Same as normal operation except at four locations

The majority of parameters measured in the network are sampled at a 0.5-s frequency and archived on a minute and hour averaged basis.

A schematic view of the elements of the RAPS air quality simulation model verification program is shown in Figure 3. Since the verification of every urban AQSM currently in existence is an unrealistic task, a representative sample of modeling techniques was selected from the modeling stockpile. The selected models will then be adapted to the St. Louis region after a 3-d period for model debugging and testing. The EPA will then use the models for the verification study.

Days are selected from the RAPS data base to form a subset of verification days to perform the studies via a series of filtering programs. The filtering criteria include such tests as

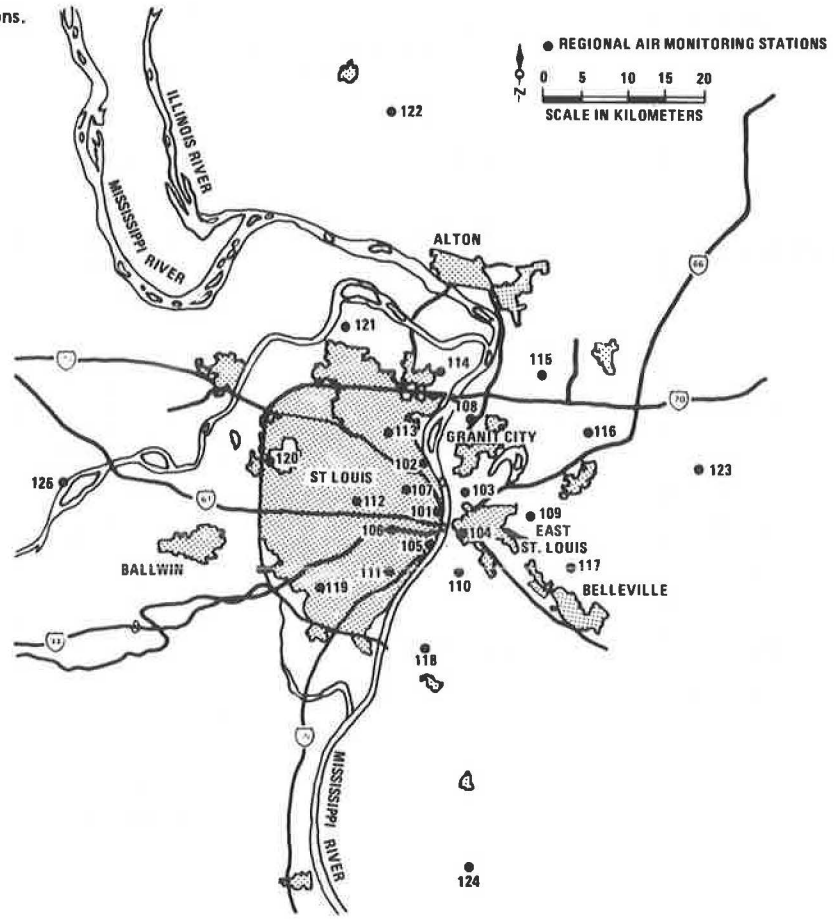
1. Days in which criteria pollutant X had Y percent or greater valid data,
2. Days in which criteria pollutant X exceeded an hourly averaged value of Y $\mu g/m^3$, and
3. Days in which criteria pollutant X exceeded a 24-h average value of Y $\mu g/m^3$.

The specific numbers used by the filters are subject to the primary pollutant to be modeled. The subset of verification days is then stratified by season. Nonreactive models will be tested against randomly selected days within each season. An attempt will be made to distribute the verification test days proportionately among the four seasons. The reactive models will use randomly selected days, but a disproportionate number of days will be selected from the summer months; fewer samples are taken in the other seasons.

After the selection of the model verification test days, a 5-d subset will be chosen and input data to the models for those days prepared. The 5-d subset will be used for preliminary testing and should allow identification of any major model inconsistencies. Obvious minor model refinements will be considered at this stage. All of the models selected will have undergone some form of evaluation and testing, so major unresolvable inconsistencies are not expected. Should such a case arise, the verification studies for the particular model in question would stop and the preliminary test results sent to the model developer for study.

Once the preliminary model testing phase is completed, the statistical verification studies will begin. Approximately 50 d of computer simulation will be considered for each model. Comparisons between computer-predicted and observed hourly averaged concentrations of pollutants will be made on a day-by-day basis. Specific statistical tests for model verification studies have been discussed by Brier (17), Nappo (18), and Liu and others (19). In light of these studies, several ap-

Figure 2. Geographical locations of RAMS stations.



proaches for the comparative studies are under consideration. Model verification will provide the first adequate set of statistical criteria on which to judge model performance and will, in the final analysis, provide the necessary information for assessment and selection of models for various application purposes.

APPLICATION AND FUTURE DEVELOPMENTS

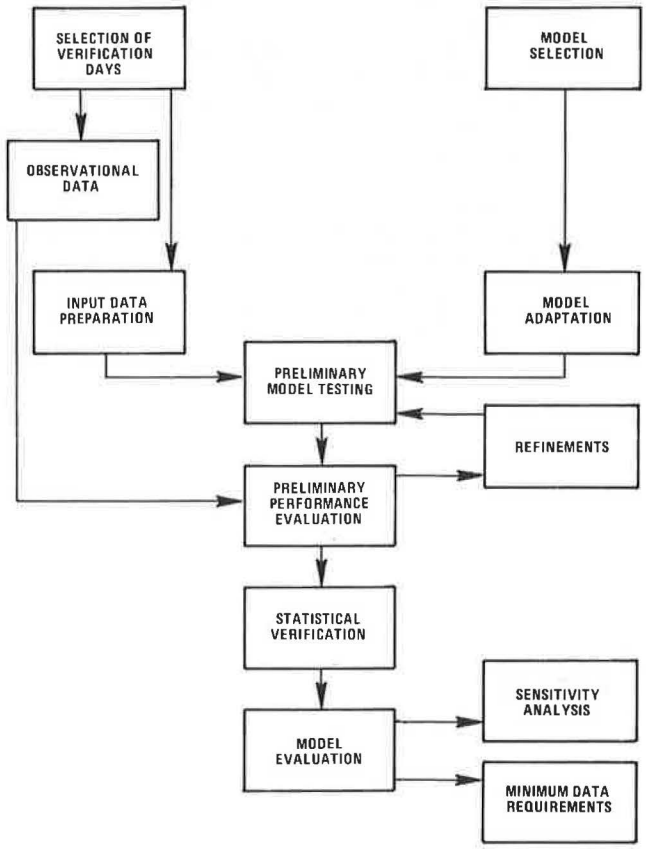
In addition to their obvious role in providing a better understanding of the fundamental chemical and physical processes associated with air pollution, PAQSMs have the potential for addressing many policy, planning, and enforcement questions in air pollution control. These include

1. Evaluation of the effectiveness of current and future emission control regulations in achieving ambient air quality standards,
2. Identification of major sources contributing significantly to air quality deterioration,
3. Determination of air quality impacts based on al-

Table 3. RAMS remote stations instrument distribution.

	Station Number																							
Parameters	101	102	103	104	105	106	107	108	109	110	111	112	113	114	115	116	117	118	119	120	121	122	123	124	125
O_3 monitor laboratories 8410	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X
NO-NO_x monitor laboratories 8440	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X
CO-CH_4-THC Beckman 6800	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X
TS-SO_2-H_2S tracor 270HA	X		X	X	X	X		X					X	X	X	X				X	X	X			
TS-Meloy SA 185		X					X		X	X	X	X					X	X	X				X	X	X
Visibility-MRI 1561	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X
Wind speed-MRI 1022 S	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X
Wind direction-MRI 1022 D	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X
Temperature-MRI 840-1	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X
Dew point-Cambridge 880	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X
Temperature gradient-MRI 840-2	X	X		X	X	X	X		X		X	X	X									X	X		
Barometer-Sostman 363	X								X			X										X	X	X	X
Solar pyranometer			X	X				X						X				X				X			
Radiation pyrheliometer			X											X				X				X			
Pyrgeometer (Eppley)			X											X				X				X			
Turbulence-R. M. Young 27002					X		X		X		X		X												
Gas bags-Xonics	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X	X
Hi-Vol-Sierra 305			X		X	X		X				X			X			X		X		X		X	
Lawrence Berkley Laboratory dichotomous sampler			X		X	X		X				X			X			X		X		X		X	
10-meter tower								X		X				X	X	X	X	X			X				
30-meter tower	X	X	X	X	X	X	X		X		X	X	X						X	X		X	X	X	X

Figure 3. Elements of the RAPS verification program for AQSM.



ternative plans for transportation and land use development, and

4. Evaluation of optimum siting of instrument stations for air quality monitoring networks.

The establishment of statistical performance criteria for AQSMs is essential if effective and creditable applications of these models are to be realized. Such performance criteria are an expected product of the RAPS model verification program. Performance criteria will vary as a function of the modeling approach as well as of the application to be considered. Therefore, the most cost-effective choice of a model for any given study must consider these three factors. Studies that assess minimum data base requirements relative to acceptable model performance must also be considered and guidance given as to the spatial and temporal resolution of model input data required. In addition, model sensitivity analyses that identify key parameters within the model are essential in providing effective use of resources in gathering data for model application. All of these studies are the subject of current and future development over the next several years.

Research and development of a regional PAQSM began early in FY 1977, under EPA sponsorship. Phase 1 of the program, scheduled for completion in early 1978, includes the basic research and formulation of the modeling framework. Phase 2, scheduled to end in early 1979, will test and evaluate the model using data gathered under the Northeast Oxidant Study Field Program of 1975. Further testing using field data gathered in other regions of the country will be considered if the phase 2 results look promising. Ultimately, the model will be used to assess impacts of air pollution control plans on pollutant concentrations many kilometers downwind. As the results from the RAPS model verification program and regional PAQSM development program unfold, further research and development studies may be indicated. The advantage will be that now the areas for refinement will be pinpointed based on extensive comparisons with observed atmospheric data.

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Regional Air Quality Modeling in Los Angeles

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The California Department of Transportation has applied regional air quality models to the Los Angeles area on a limited basis. A Lagrangian, or trajectory, model was used to evaluate the air quality impact of a highway project. An Eulerian, or grid, model is currently being used to evaluate regional transportation plans. The use of these models requires a costly and extensive data base for validation and the cooperation of local and regional agencies. A large aerometric sampling program was undertaken for one of the data bases. Technical problems in model application were related to electronic data processing and reconciliation of data that are tied to different geographic grid systems. In-house computer expertise is essential for application of the model. Model output tends to be voluminous and requires simplification for analysis and presentation.

The Los Angeles region is reputed to have one of the world's most intractable oxidant problems. In June 1974, for example, a 5-d oxidant episode created a peak 1-h value of about 860 $\mu g/m^3$ (44 pphm) in the eastern portion of the Los Angeles basin. The national ambient air quality standard (NAAQS) of 160 $\mu g/m^3$ (8 pphm) was violated for 1400 h that year. In years prior to 1974, even higher values of oxidant have been observed.

The Los Angeles Federal Air Quality Control Region (LAAQCR) encompasses all parts of the six counties shown in Figure 1. This region is roughly 129 km (80 miles) long and 257 km (160 miles) wide and includes approximately 25 205 km^2 (9736 $miles^2$) of land and a population of 10 million people. The principal agency for overall planning in the region is the Southern California Association of Governments (SCAG). This agency is supported by the 127 member cities and five counties within the region and by the California Department of Transportation (Caltrans).

The air quality management planning effort is funded by the U.S. Environmental Protection Agency (EPA). The funds are channeled through the California Air Resources Board (CARB) to the Air Quality Maintenance Planning (AQMP) group, which is also supported by the staffs of the CARB, SCAG, the four-county Southern California Air Quality Management District (SCAQMD), the Ventura County Air Pollution Control District (VCAPCD), the Santa Barbara County Air Pollution Control District (SBAPCD), and Caltrans.

The LAAQCR is a plain, bounded on the north by a high mountain range and on the west by the Pacific Ocean. Some internal geographic features divide it into subregions but do not usually block the west-to-east movement of air through the basin. Typical meteorology during an oxidant episode is a west-to-east air movement at moderate speed, a subsidence inversion producing a mixing depth of 300 to 460 m (1000 to 1500 ft), clear skies, and high ambient temperature. During the night, a wind reversal produces a sloshing effect. These conditions occur several times during the oxidant season (May to October). Other weather patterns (such as weak Santa Ana winds) also produce episodes, but this pattern typically produces the highest levels of oxidants.

The LAAQCR has 44 stations that measure air quality and many that measure wind speed and direction. Much of these data must be used with caution as the wind instruments at the measuring stations are not necessarily sited to produce wind data that are representative of the surrounding area. The sources of pollution are highly variable and geographically dispersed. The CARB emission inventory for the region estimated a background level (geogenic origin) of 1.36×10^5 kg/d (150 tons/d) of reactive hydrocarbons (RHC). In 1975, the daily production of RHC from anthropogenic sources was about 1.41×10^6 kg/d (1550 tons/d). This was generated 40 percent from light-duty vehicles, 20 percent from all other forms of transportation, and 40 percent from industrial and area sources. Much of the RHC is the result of industrial activities in the western part of