# DEVELOPMENT OF A SIMULATION MODEL FOR ESTIMATING GROUND LEVEL CONCENTRATIONS OF PHOTOCHEMICAL POLLUTANTS

FINAL REPORT

Prepared by

Systems Applications, Inc. Beverly Hills, California 90212

for the

Air Pollution Control Office of the Environmental Protection Agency Durham, North Carolina 27701

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#### ABSTRACT

In this report we describe the progress that has been achieved to date in the development and validation of a simulation model for estimating ground level concentrations of photochemical pollutants. This model is based on the finite difference solution of the equations of conservation of mass, using the method of fractional steps. The bulk of the effort reported here is developmental, involving the compilation of a comprehensive source emissions inventory, the development and validation of a kinetic mechanism for photochemical reactions, the adaptation of the method of fractional steps for use in the solution of the governing equations, and the preparation of maps displaying spatial and temporal variations in wind speed and direction and in the height of the inversion base. The details of these various efforts are described in a series of appendices to this report. Although a validated kinetic mechanism has been developed and incorporated in the simulation model, validation efforts have thus far been restricted to carbon monoxide. Provisional validation results for the Los Angeles Basin are presented.

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# INTRODUCTION

Urban airshed models are mathematical representations of atmospheric transport and chemical reaction processes which, when combined with a source emissions inventory and pertinent meteorological data, may be used to predict pollutant concentrations at any point in the airshed. Models capable of accurate prediction will serve as an important aid in urban and regional planning. In particular, these models will be used for:

- Simulation of the effects of alternative air pollution control strategies on pollutant concentrations in the airshed
- Planning for land use, so that projected freeways, industrial sites, and power plants may be located where their air pollution potential is minimized
- Determination of the long-term air pollution control strategy which accomplishes desired air quality objectives at least cost
- Real-time prediction in an alert warning system, such that an impending air pollution episode may be anticipated and proper preventive action taken.

An airshed simulation model that is to be generally useful in urban planning studies must meet several requirements.

- It should be capable of predicting accurately the ground level concentrations of inert pollutants, as well as those formed in the atmosphere through chemical reactions. Of immediate interest, and the focus of this study, is the prediction of carbon monoxide, nitrogen oxide, ozone, and hydrocarbon concentrations and their variations in space and time. Also of interest, particularly for large Eastern and Midwestern cities, is the estimation of sulfur dioxide and particulate concentrations. Finally, it should be possible to incorporate into the model, when the means are developed, the facility for estimating aerosol concentrations.
- The model should have a spatial and temporal resolution appropriate for the analysis of concentration variations which occur in a city throughout the course of day. For a typical large urban area, the horizontal spatial resolution may be of the order of a mile, and the temporal resolution, of the order of an hour. The resolution of the model will, of course, be influenced by the availability of data of similar resolution.
- The complexity of the model, and thus the computing time and computer storage requirements, should be such that the model can be operated at a reasonable cost using computers of general availability.

Models that have been developed in the past, and that have had wide application, fail to satisfy all these requirements.

The simplest of the existing models is the so-called "box model", wherein pollutant concentrations are assumed to be homogeneous throughout the entire airshed. In addition, it is assumed that, within the airshed:

- The sources are distributed uniformly
- Emitted pollutants are instantaneously and uniformly mixed
- A uniform wind characterizes transport
- A constant inversion height is typical of time-averaged meteorology

A model of this type has been employed by Smith (1961) to estimate pollutant concentrations in an urban area. A variant of the box model, consisting of a two-dimensional network of interconnected boxes or well-mixed cells, has been developed by Reiquam (1970a, 1970b, 1970c) to estimate monthly average pollutant concentrations in the Willamette Valley and in Northern Europe. In this model, the assumptions listed above apply to each well-mixed cell. The highly simplified box model clearly lacks the spatial resolution needed to properly represent the distribution of sources or to estimate spatial concentration variations in an urban airshed. However, its variant, the well-mixed cell model, does not suffer this deficiency. In fact, Reiquam's formulation can be extended for application to the prediction of pollutant concentrations in urban areas and to include photochemical reactions. The well-mixed cell model is further discussed in Section I.B.3.

The second type of model that has been applied in the prediction of pollutant concentrations is the plume model. This model was originally developed to describe the concentration distribution of an inert species downwind of a point source. It has subsequently been extended for application to line and area sources and, by imposing the principle of superposition, to a distributed array of sources. In the usual applications of this model:

- Only inert pollutants are considered
- Wind shear is neglected
- Measures of plume spread are assumed constant, are based on experimental studies (usually carried out over rural areas), are independent of height, and are a function of atmospheric stability class.

While the plume model has been widely applied during the past decade in the prediction of concentrations of sulfur dioxide and particulates in urban areas (see, for example, Turner (1964), Clarke (1964), Miller and Holzworth (1967), Koogler, et al. (1967), Hilst (1967), Slade (1967), and Bowne (1969)), it is of little use in the prediction of concentrations of pollutants formed through chemical reaction. Nevertheless, gaussian plume models can often provide useful estimates of concentrations downwind of strong isolated sources.

Reviews of gaussian plume and puff models\* have been presented by Lamb (1968), Seinfeld (1970), and Neiburger and Chin (1970).

As models of a relatively simple nature are inadequate to meet the requirements stated, we consider next more fundamental approaches to the simulation of transport and reaction processes. The most complex of these involves the solution of the turbulent planetary boundary layer equations for the conservation of mass, momentum, and energy. The solution of these equations is a truly prodigious undertaking, the demands of which exceed the computing speeds and storage capacities of the present generation of computers. Less ambitious is the solution of one (or possibly two) of these three classes of equations, at the same time supplying as data the information that would not be computed by the model due to the omission of one or more classes of equations from the system description.

One such approach to airshed modeling is the solution of the partial differential equations of conservation of mass. This approach has appeal because it provides a means for including chemical reaction phenomena, time-varying meteorological conditions, and complex source emissions patterns while avoiding an undue level of complexity. However, the computational requirements for the solution of these equations are still substantial, both with respect to computing time and computer storage. Data requirements are also considerable, as those variables associated with the momentum and energy equations, such as wind speed and direction and height of the inversion base, must be treated as inputs to the model. Thus, the questions that must be answered in the development of such a model are:

- 1) Will it predict ground level concentrations of primary and secondary pollutants\*\* with acceptable accuracy?
  - 2) Assuming that the model is of acceptable accuracy, can it be operated at a reasonable cost, given the large computing time and computer storage requirements associated with such an effort?

It has been the purpose of this study to develop and validate an airshed simulation model based on the solution of the equations of conservation of mass, and, in so doing, to seek answers to these questions. In this report we detail the progress that has been made to date in this effort. In Section I, we review the various approaches to airshed modeling that are based on the solution of the continuity equations. We comment on the virtues and deficiencies inherent to each approach, and present a comparative evaluation. In Section II, we describe in more detail the particular approach adopted in this study. Progress to date in the validation of this model is summarized in Section III, and we conclude, in Section IV, with an outline of recommended future efforts.

<sup>\*</sup> The puff model, described by Roberts, et al. (1970), is an extension of the plume model, in that it is based on the gaussian distribution and the same estimation procedures for dispersion parameters. However, by assuming that emissions can be treated as discrete puffs, certain assumptions normally made for the plume model can be relaxed, notably that of steady state behavior., However, chemical reactions have yet to be treated in this approach.

\*\*Primary pollutants are those that do not undergo, or have not undergone, chemical change subsequent to being emitted. Secondary pollutants are those species that are formed in the atmosphere through chemical reaction processes.

# I. AIRSHED MODELS BASED ON THE EQUATIONS OF CONSERVATION OF MASS

There are several approaches to airshed modeling based on the solution of the equations of conservation of mass.\* These may be divided into two basic categories:

- Moving cell models
- Fixed coordinate models

In the moving cell approach a hypothetical column of air, which may or may not be well-mixed vertically, is followed through the airshed as it is advected by the wind. Pollutants are injected into the column at its base and chemical reactions may take place within the column. In the fixed coordinate approach the airshed is divided into a three-dimensional grid, which can be envisioned as stacked layers of cells, each cell being perhaps one to two miles on a side and of the order of one hundred feet high. This three-dimensional grid is then used as a basis for the numerical solution of the equations of conservation of mass. In the language of fluid mechanics, the moving cell approach is termed Lagrangian, and the fixed coordinate approach, Eulerian. Each of the approaches has characteristics which suggest application in the analysis of particular types of air pollution problems.

The modeling approach we have adopted and pursued in our study is Eulerian in nature and is based on the finite difference solution of the equations of conservation of mass. This approach, as do all others involving the solution of the continuity equations, requires several components:

- A kinetic mechanism describing the rates of atmospheric chemical reactions as a function of the concentrations of the various species present.
- A source description, giving the temporal and spatial distribution of emissions from all significant pollutant sources in the airshed.
- A meteorological description, including wind speed and direction at each location in the airshed as a function of time, the vertical atmospheric temperature profile and radiation intensity.

But it is to the overall model, in which these components are embedded, that we must first direct our attention. For it is the nature and structure of this model that will determine the degree to which the requirements that are placed on an airshed model will be met.

In this section, we review the various approaches that are currently being pursued in the simulation of atmospheric transport and reaction processes which are based on the solution of the continuity equations. We

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<sup>\*</sup> Recall that we have restricted this discussion to models capable of describing concentration changes in an urban airshed over time intervals of the order of a day. We have also excluded from consideration urban air pollution analyses of restricted scale, such as the prediction of concentrations in the vicinity of major local sources, notably freeways, airports, power plants and refineries.

comment on the virtues and deficiencies inherent to each, and at the conclusion present a comparative assessment. It is hoped that, as a result of this review, the approach we have adopted can be better understood and evaluated when compared with other modeling efforts under development. We begin the discussion by presenting the equations upon which all models to be described are based.

The time-averaged equation of conservation of mass for species i in a turbulent flow is given by (see Bird, et al. (1960)):

$$\frac{\partial \overline{c_i}}{\partial t} + \frac{\partial (\overline{uc_i})}{\partial x} + \frac{\partial (\overline{vc_i})}{\partial y} + \frac{\partial (\overline{wc_i})}{\partial z} + \frac{\partial (\overline{u'c_i'})}{\partial x} + \frac{\partial (\overline{v'c_i'})}{\partial y} + \frac{\partial (\overline{w'c_i'})}{\partial z}$$

$$= D_i \left( \frac{\partial^2 \overline{c_i}}{\partial x^2} + \frac{\partial^2 \overline{c_i}}{\partial y^2} + \frac{\partial^2 \overline{c_i}}{\partial z^2} \right) + R_i (\overline{c_i} + c_i') + C_i' + C_i$$

where u, v, w = time-averaged wind velocity components in the x, y and z directions respectively

u',v',w' = instantaneous turbulent velocity fluctuations

c<sub>4</sub> = time-averaged concentration of species i

c; = turbulent concentration fluctuation of species i

D, = molecular diffusivity of species i in air

 $R_{i}$  = rate of generation of species i by chemical reaction

S, = rate of emission of species i by elevated sources

- If: A. Molecular diffusion is negligible when compared with turbulent diffusion
  - b. The contribution of turbulent concentration fluctuations to  $\mathbf{R}_{i}$  are neglected

c. The flow is incompressible, i.e., 
$$\frac{\partial u}{\partial x} + \frac{\partial v}{\partial y} + \frac{\partial w}{\partial z} = 0$$

d. Turbulent eddy diffusivities are defined by:

$$\frac{\mathbf{u'c'_1}}{\mathbf{v'c'_1}} = -K_{\mathbf{x}} \frac{\partial \mathbf{c'_1}}{\partial \mathbf{x}}$$

$$\frac{\partial \mathbf{c'_1}}{\partial \mathbf{x'c'_1}} = -K_{\mathbf{x}} \frac{\partial \mathbf{c'_1}}{\partial \mathbf{x'}}$$

then Equation (1) simplifies to

$$\frac{\partial \overline{c}_{i}}{\partial t} + \overline{u} \frac{\partial \overline{c}_{i}}{\partial x} + \overline{v} \frac{\partial \overline{c}_{i}}{\partial y} + \overline{w} \frac{\partial \overline{c}_{i}}{\partial z} = \frac{\partial}{\partial x} \left( K_{x} \frac{\partial \overline{c}_{i}}{\partial x} \right) + \frac{\partial}{\partial y} \left( K_{y} \frac{\partial \overline{c}_{i}}{\partial y} \right) + \frac{\partial}{\partial z} \left( K_{z} \frac{\partial \overline{c}_{i}}{\partial z} \right) + R_{i} (\overline{c}_{1}, \dots, \overline{c}_{p}) + S_{i} \qquad i = 1, 2, \dots, p$$
(2)

These p coupled equations provide a convenient basis from which to describe both the moving cell and fixed coordinate approaches. It should be noted that the coupling of the equations occurs through the nonlinear chemical reaction term  $\hat{R}_{i}$ . The functional form of  $R_{i}$  derives from the particular formulation upon which the kinetic mechanism for the chemical reactions is based (see Appendix C).

# A. Moving Cell Approaches

As we have noted, the principal feature of the moving cell approach is that concentration changes in a hypothetical parcel of air are computed as the parcel traverses the airshed. The parcel is visualized as a vertical column of air of fixed cross-sectional area and variable height, with the top of the column being defined by the base of an elevated inversion or, in the absence of an inversion, by an estimated maximum mixing height. The motion of the air column is assumed to correspond to the local instantaneous wind speed and direction, thereby tracing out a particular surface trajectory in the airshed.

The following assumptions are inherent in this model:

- There is no horizontal transport of material across the boundaries of the parcel (there is, in fact, no means of including horizontal diffusive transport between the column and the environment).
- There is no change in the horizontal wind velocity with height.
- Vertical advection is neglected, i.e., w = 0.

The basic assumption underlying the approach is that a parcel of air maintains its integrity while traversing the airshed. It is highly unlikely that this is ever the case in the atmosphere over the time scales of interest.

Since horizontal transport across the boundaries of the column is neglected, and since the column moves with the average ground-level horizontal wind velocity, the moving cell approach may be represented rather simply mathematically. If the contents of the column are considered to be horizontally uniform but vertically non-uniform, the only independent variables are time t and vertical distance z. The concentration of species i,  $\overline{c}_i(z,t)$ , is determined by integration of the abridged form of Equations (2):

$$\frac{\partial \overline{c}_{i}}{\partial t} = \frac{\partial}{\partial z} \left( K_{z} \frac{\partial \overline{c}_{i}}{\partial z} \right) + R_{i} (\overline{c}_{i}, \dots, \overline{c}_{p}) + S_{i}$$
(3)

$$i = 1, 2, ..., p$$

The initial condition for this formulation is that the concentration within the column at the beginning of the traversal be given, i.e.,

$$\overline{c}_{i}$$
 (z,0) =  $\overline{c}_{i}$ 

The boundary conditions at the ground, z = 0, and at the inversion base (or top of the column), z = H(t), are given by

$$-K_{z} \frac{\partial c_{i}}{\partial z} = Q_{i}(t) \qquad z = 0$$

$$-K_{z} \frac{\partial c_{i}}{\partial z} = 0 \qquad z = H(t)$$

where  $Q_i(t)$  is the flux of species i from ground-level sources,  $S_i$  is the mass rate of emission of species i from elevated sources, and H(t) is the height of the column as a function of time. The movement of the column is reflected mathematically only in  $Q_i(t)$  and H(t). An approach based on Equations (3) has been developed for Los Angeles by Eschenroeder and Martinez (1969).

A simplified version of this model results if we neglect vertical inhomogeneities in the column. Then  $\overline{c}_i = \overline{c}_i(t)$  only, and Equations (3) reduce to

$$\frac{d(\overline{Vc_i})}{dt} = VR_i(\overline{c_1}, \dots, \overline{c_p}) + AQ_i + VS_i$$
 (4)

where the column is simply a well-mixed vessel having a volumn V, a base area A, and a time varying pollutant input rate  $AQ_1 + VS_1$ . The advantage of this approach, when compared to that based on Equations (3), is mainly ease of computation, since Equations (4) consist of ordinary rather than partial differential equations. Wayne, et al. (1971) have utilized this approach in developing a model of the Los Angeles Basin.

The moving cell approach has the following virtues:

- The lengthy integration of Equations (2) in all three spatial dimensions and time is avoided.
- The concentration history along an air trajectory can be traced, thereby permitting an assessment of the effect of specific sources at locations downwind of these sources.

# and deficiencies:

- The concept of an identifiable parcel of air is an oversimplification since such an entity never exists in a turbulent atmosphere over time scales of interest.
- There is no way to include convergence and divergence phenomena in the wind field, and the resulting vertical advection of air.
- In order to determine the concentration at a given location and time, it is necessary to trace the trajectory backward in time to the point where it entered the airshed. Since the only reason for this calculation is to ascertain the starting-point of the trajectory, its inclusion constitutes an inefficiency inherent in the approach, particularly when a large number of trajectories must be computed.

One final observation. While the moving cell approach is a useful technique for computing concentration histories along a given air trajectory, it is not practical for full airshed modeling. This is largely because of the great many trajectory calculations required to construct maps of predicted concentrations for a wide area.

# B. Fixed Coordinate Approaches

In the fixed coordinate approach to airshed modeling, the airshed is divided into a three-dimensional grid for the numerical solution of some form of Equations (2), the specific form being dependent upon the simplifying assumptions made. We can classify the general methods for solution of the continuity equations as:

- Conventional finite difference methods
- · Particle-in-cell methods
- Variational methods

We will discuss in this section finite difference methods and particlein-cell methods. Variational methods (of which a specific class, Galerkin methods, are particularly pertinent) involve assuming the form of the concentration distribution, usually in terms of an expansion of known functions, and then evaluating coefficients in the expansion. There is currently very active interest in the development of these techniques (see, for example, Douglas and DuPont (1970)); however, they are not included in this discussion, as experience to date in their application to complex systems of differential equations is very limited.

The principal numerical problems associated with fixed coordinate methods are that:

- Complex and lengthy calculations are required for the integration of several coupled nonlinear partial differential equations in three dimensions.
- Changes in elevation of the upper boundary (i.e., H(x,y,t)) with time require repeated reconfiguration of the grid on which the solution is carried out.

However, models based on a fixed coordinate approach may be used to predict pollutant concentrations at all points of interest in the airshed at any time. This is in contrast to the moving cell methods, wherein predictions are confined to the paths along which concentration histories are computed.

# 1. Finite-Difference Methods

The numerical analysis literature abounds with finite-difference methods for the numerical solution of partial differential equations. Unfortunately, while these methods have been successfully applied in the solution of two-dimensional problems in fluid mechanics and diffusion (see, for example, Peaceman and Rachford (1962) and Burstein (1967)). There is a dearth of reported experience in the solution of three-dimensional, time-dependent, nonlinear problems. Application of these techniques, then, must proceed by extending methods successfully applied in two-dimensional formulations to the more complex problem of solving Equations (2). For general discussions of the various types of finite-difference methods applicable in the solution of partial differential equations, and their advantages and disadvantages, we refer the reader to the books by von Rosenberg (1969), Forsythe and Wasow (1960), and Ames (1969).

The principal considerations in choosing a finite-difference method for the solution of the continuity equations are accuracy, stability, computation time, and computer storage requirements. Accuracy of a method refers to the degree to which the numerically computed temporal and spatial derivatives approximate the true derivatives. Stability considerations place restrictions on the maximum time step  $\Delta t$  that can be used in the integration. Implicit methods, those involving the simultaneous solution of difference equations at each step, are more suitable for the solution of nonlinear forms of the continuity equations than are explicit methods, as the former are stable over a wider range of step sizes. Implicit methods, however, involve considerably more computation per time step than do explicit methods. Other finite difference methods exist which are difficult to classify.

Typically, these techniques have the characteristics of implicit methods,

yet, because of some unique aspect of the particular method, involve less burdensome calculations than are normally expected with an implicit method. Two such techniques that have the potential for application in the solution of Equation (2) are the method of fractional steps and the method of alternating directions (see Richtmyer and Morton (1967) for a discussion of these techniques).

To date, there have been reported only two applications of finite-difference methods to the solution of the equations of conservation of mass as they pertain to urban airsheds, both for the Los Angeles Basin. These are the work of Eschenroeder and Martinez (1969) and that described in this report. Eschenroeder and Martinez applied the Crank-Nicolson implicit method to the simplified version of Equations (2),

$$\frac{\partial \overline{c_1}}{\partial t} + \overline{u} \frac{\partial \overline{c_1}}{\partial x} = \frac{\partial}{\partial z} \left( K_z \frac{\partial \overline{c_1}}{\partial z} \right) + R_1$$

In a later paper (Eschenroeder and Martinez (1971)), they report that a number of difficulties were encountered in using the Crank-Nicolson method and the approach was abandoned. In the work we have carried out, the method of fractional steps has been applied to the solution of six equations of the form of Equations (2), four of which are coupled. (While horizontal dispersion was neglected in the final formulation, it was originally incorporated through the use of Equation (6)). This work is described in Sections II and III and in Appendix D.

The main advantage in using a finite-difference method in the solution of Equations (2), as compared with other fixed coordinate approaches, is that there has been extensive experience in applying these methods to a wide variety of partial differential equations. Even though reported experience with three-dimensional, time-dependent, nonlinear problems is scanty, experience with simpler systems provides a sound basis for the development of feasible approaches. The disadvantages of finite-difference methods are well known:

- Inaccuracies in approximating the first-order advection terms in the continuity equations give rise to errors which have the mathematical characteristics of diffusion processes. These inaccuracies, termed "numerical" or "artificial" diffusion, often mask the representation of true diffusion.
- Computing time and storage requirements associated with accurate, stable methods can be excessive for problems involving several independent variables. (This is also true, however, for all fixed coordinate methods.) When the equations are nonlinear, time-consuming iterations or matrix inversions are often required in their solution.

# 22. Particle-in-Cell Methods

An alternative to the direct finite-difference solution of Equations (2) is the so-called particle-in-cell (PIC) technique. The distinguishing feature of the PIC technique is that the continuous concentration field is treated as a collection of mass points, each representing a given amount of pollutant and each located at the center of mass of the volume of material it represents. The mass points, or particles, are moved by advection and diffusion. It is convenient, but not necessary, to have each of the particles of a given contaminant represent the same mass of material. The application of the PIC technique in hydrodynamic calculations is discussed by Harlow (1964). We consider here the use of the PIC technique in the numerical solution of Equations (2).

Given an initial, continuous concentration field in the airshed, we replace this field by discrete particles of pollutant i, each representing a fixed mass. The particles are located within a three-dimensional fixed grid according to the mass distribution of material. Thus, each particle has a given set of coordinates. Consider now a single time step  $\Delta t$  in the numerical solution of the continuity equations using the PIC method. We write Equations (2) in the form

$$\frac{\partial c_1}{\partial t} + \nabla \cdot \overline{v_1} c_1 = R_1 + S_1$$
 (5)

where the effective velocity  $\overline{\mathbf{U}}_{\mathbf{i}}$  is defined by

$$\overline{\overline{v}}_{i} = \overline{\overline{v}} - \frac{\overline{x}}{\overline{\varepsilon}_{i}} \quad \overline{\nabla c}_{i}$$
 (6)

$$\overline{\underline{\mathbf{v}}}_{\mathbf{x}} = \begin{bmatrix} \mathbf{v}_{\mathbf{x}} \\ \mathbf{v}_{\mathbf{x}} \end{bmatrix}$$
 and  $\mathbf{K} = \begin{bmatrix} \mathbf{k}_{\mathbf{x}} \\ \mathbf{k}_{\mathbf{z}} \end{bmatrix}$ . In the computational procedure,

each particle of Species i at location (x,y,z) is moved a distance  $|\overline{y}|\Delta t|$  in the direction of  $\overline{y}|\Delta t|$ . In addition, new particles are emitted during the period t to t +  $\Delta t$  from the sources located in each cell, the number of particles emitted being determined by the product of the source strength and the time step. These new particles are also advected with velocity  $\overline{y}|_{\bullet}$ . After the convective step the average concentration of each species in a cell is calculated, this concentration being equal to the total mass of particles occupying the cell, divided by the cell volume. The cell contents are then allowed to react, resulting in a concentration change,  $R_{\bullet}\Delta t$ . Finally, the particles are reconstituted, with the change in mass due to chemical reaction being reflected in changes in the number of particles of each species. The same procedure is repeated for succeeding time steps. The PIC technique has been adapted to air pollution modeling by Sklarew (1970a, 1970b).

The PIC technique has the following advantages:

 Artificial diffusion due to truncation errors in the advection terms in Equations (2) is eliminated since these terms are not approximated by finite-difference representations.

- There are no stability restrictions on  $\Delta t$  (although  $\Delta t$  should be small enough so that the value of  $\overline{U}_i$  is representative of the movement of fluid particles).
- Particles can be tagged as to their place of origin, thus making it possible to identify the sources of contaminants observed at any location.

# and weaknesses:

- Computer storage requirements can become excessive, as the coordinates of a large number of particles must be kept in memory.
- . If it is assumed that each particle of a given contaminant represents the same mass of material, then every cell will have a residue that cannot be assigned to a particle. On the average, this residual material will equal one-half of a particle mass. For example, for the simple case in which there are four particles in every cell, then, on the average, ll% of the total material will not be included in particles.

#### 3. Well-Mixed Cell Model

A conceptually simple approach is based on the representation of the airshed by a three-dimensional network of well-mixed vessels (see Seinfeld (1970)). As before, we assume that the airshed has been divided into an array of L cells. Instead of using the array simply as a tool in the finite-difference solution of the continuity equations, let us now assume that each of these cells is actually a well-mixed reactor with inflows and outflows between adjacent cells. If we neglect diffusive transport across the boundaries of the cells and consider only convective transport among cells, a mass balance on species i in cell k is given by

$$v_{k} \frac{d\overrightarrow{c}_{ik}}{dt} = -\overrightarrow{c}_{ik} \frac{dv_{k}}{dt} + \sum_{j=0}^{L} q_{jk} \overrightarrow{c}_{ij}$$

$$-\overrightarrow{c}_{ik} \sum_{j=0}^{L} q_{kj} + s_{ik} + R_{i} (\overrightarrow{c}_{1k}, \dots, \overrightarrow{c}_{pk})$$
(7)

where

c<sub>ik</sub> = concentration of species i in cell k

 $V_k$  = volume of cell k

q jk = volumetric flow rate of air from cell j to cell k,
 q is the flow from the exterior of the airshed into
 cell k, and q is the flow from cell k to the exterior
 of the airshed

S<sub>ik</sub> = rate of emission of species i into cell k from all sources

 $R_{i}$  = rate of formation of species i by chemical reaction

Normally,  $dv_k/dt$  is set equal to  $A_k(dH_k/dt)$ , where  $A_k$  is the area of the base of a cell having vertical sides and  $H_k$  is the height of the top of the cell. In effect, the cell is a box with permeable walls and a movable lid.

If we divide the airshed into L cells and consider p species, Ip ordinary differential equations of the form of Equations (7) constitute the airshed model. As might be expected, this model bears a direct relation to the partial differential equations of conservation of mass. If we allow the cell size to become small, it can be shown that Equations (7) are the same as the first-order spatial finite difference representation of Equations (2) in which turbulent diffusive transport is neglected, namely

$$\frac{\partial \overline{c_i}}{\partial t} + \overline{u} \frac{\partial \overline{c_i}}{\partial x} + \overline{v} \frac{\partial \overline{c_i}}{\partial y} = R_i + S_i$$

Therefore, the well-mixed cell model can also be described as the result of the finite difference approximation of the spatial derivations of (2), that is, of the conservation equations in which diffusion has been neglected.

The advantages of the well-mixed cell approach are as follows:

- The geometries of cell bases (which may be both irregular and variable from cell to cell) can be drawn to conform with topographic features.
- Variations in inversion height with time are easily incorporated in the model.
- The model is conceptually easy to understand and implement (only ordinary differential equations are involved).

Its disadvantages, however, are considerable.

• Due to the large variations that can occur in the magnitudes of the flows, q<sub>jk</sub>, Equations (7) are often "stiff", thus requiring implicit integration techniques to insure stability in their solution.\* If an implicit technique is used, the inversion of an Ip x Ip matrix is necessary at each time step. Since computing the inverse of large matrices can be very time-consuming, this requirement places a definite restriction of the size of L. For example, if we were to consider 25 cells for our system of four coupled equations, the repeated inversion of a 100 x 100 matrix would be required.

<sup>\*</sup>See Appendix B, Section IV, for a discussion of the solution of "stiff" systems of ordinary differential equations.

- Diffusive transport is neglected. This is a distinct drawback in the case of vertical diffusion.
- The mathematical formation of the well-mixed cell model, Equations (7), is such that the expected accuracy of the solution is equivalent to that expected from the application of only a first order finite difference method to the solution of a corresponding model based on the partial differential equations, Equations (2).

# C. An Assessment

Of the four models described that are based on the equations of conservation of mass, all are at an initial stage of development. It is thus premature to evaluate them. As has been pointed out, the major distinction to be drawn among the models is that which exists between the moving cell and fixed coordinate approaches. Moving cell models have their most significant application in exploring phenomena of a restricted scope in the spatial domain, such as examining the possible sources of pollutants that are concentrated in a particular region at a given time. Thus, while these models are not intended as full airshed models, they have the advantage, when compared to fixed coordinate approaches, of computational simplicity.

In contrast, the fixed coordinate approach is readily applicable to the prediction of pollutant concentrations at all points in an airshed at any time. These predictions can serve as the basis for the preparation of contour plots of estimated concentration levels. However, the costs that must be incurred to obtain these results are high, due to the substantial computing time and storage requirements associated with this approach. Much less can be said, however, in comparing the PIC and finite difference approaches to the solution of Equations (2). Both have inherent sources of inaccuracy (the discretization of the concentration field in creating a finite number of particles in the PIC approach, the error of truncation and resulting "artificial" diffusion in finite difference approaches) that can be reduced only at a cost of substantially increasing computing time and computer storage requirements (the creation of a larger number of particles, each of a reduced mass, in the PIC approach, and the use of a finer spatial grid and a higher order finite difference method in the finite difference approach). The only means for assessing the relative advantages of these approaches, in terms of accuracy and costs incurred in their application, is through direct comparison. Models are only now reaching a state of development at which such comparative studies are feasible. As of this writing, however, there is no means for assessing the relative merits of these airshed models.

The modeling approach that we have pursued in this study has been the development of a comprehensive airshed model based on the finite difference solution of the equations of conservation of mass. In the next section, we present the details of this model development effort.

# II. THE MODEL DEVELOPED IN THIS STUDY

The airshed simulation model we have developed is based on the following formulation of the equations of conservation of mass\*:

$$\frac{\partial c_{i}}{\partial t} + u \frac{\partial c_{i}}{\partial x} + v \frac{\partial c_{i}}{\partial y} + w \frac{\partial c_{i}}{\partial z} = \frac{\partial}{\partial z} \left( K_{z} \frac{\partial c_{i}}{\partial z} \right) + R_{i}(c_{1}, c_{2}, \dots, c_{p}) + S_{i}$$

$$i = 1, 2, \dots, p$$
 (8)

for 
$$x_W \le x \le x_E$$

$$y_S \le y \le y_N$$

$$h(x,y) \le z \le h(x,y,t)$$

$$t \ge t$$

where x,y = horizontal coordinates

z = vertical coordinate

u,v,w = three components of average
 wind velocity vector

c, = time-averaged concentration of species i

K = turbulent eddy diffusivity

S<sub>i</sub> = rate of emission of species i from elevated sources

R<sub>1</sub> = rate of production of species i
 through chemical reaction

 $x_{W}$ ,  $x_{E}$ ,  $y_{S}$ ,  $y_{N}$  = west, east, south, and north boundaries

h(x,y) = terrain elevation

H(x,y,t) = elevation of the inversion base above sea level

The initial and boundary conditions are:

initial  $c_i(x,y,z,t_0) = f_i(x,y,z)$ 

boundary (1) 
$$-K_{z} \frac{\partial c_{i}}{\partial z} = Q_{i}(x,y,t) \quad \text{at } z = h(x,y)$$

<sup>\*</sup> Overbars (~) indicating averaged quantities will be omitted henceforth. All velocities and concentrations, however, continue to be time averaged quantities.

boundary (2) if 
$$W \ge 0$$
, then  $-K_z \frac{\partial c_i}{\partial z} = 0$ 

if W < 0, then W g<sub>1</sub>(x,y,z,t) = W<sub>2</sub>c<sub>1</sub> - K<sub>2</sub> 
$$\frac{\partial c_1}{\partial z}$$
 at z = H(x,y,t)

where 
$$W = w - u \frac{\partial H}{\partial x} - v \frac{\partial H}{\partial y} - \frac{\partial H}{\partial t}$$

(3) 
$$c_{\underline{i}} = g_{\underline{i}}(x,y,z,t) \qquad \text{at } x = x_{\underline{W}} \text{ (or } x_{\underline{E}})$$

$$y = y_{\underline{G}} \text{ (or } y_{\underline{N}})$$

where x and y are at boundaries through which the prevailing winds enter.

and where  $Q_i(x,y,t) = surface flux of species i$ 

In addition to the assumptions made in deriving Equations (2) in Section I, several features of the formulation should be noted:

- · Horizontal turbulent diffusion is neglected.
- Due to changes in terrain elevations with x and y, the vertical component of the wind will not, in general, be zero at the ground.
- The boundary condition at the inversion base (z = H) is the usual flux condition if the transformed vertical velocity component W is upward. If W is down into the airshed at z = H, the concentration of inflowing material is specified.

The most notable feature of the equations, however, is that they are coupled, since they share a common argument through the terms  $R_i(c_1, c_2, \ldots, c_p)$ . We are thus confronted with the problems of solving p coupled, nonlinear partial differential equations in four dimensional space (x,y,z,t).

Up to this point we have described airshed models and modeling in the abstract. When we plan to actually undertake solution of the equations, however, we must do so for a particular region. Source emissions rates and their distribution in space and time and meteorological variables, such as wind speed and direction and mixing depth, are inputs to the model. In order to test (or validate) the model, it is necessary to include as inputs the source distribution and meteorology appropriate for the area during the time period in question, and to compare the pollutant concentrations predicted with concentrations actually measured in the area during that period.

The Los Angeles Basin was the obvious choice for the prototype study, largely because of the availability of a relatively rich data base. A network of nearly three dozen wind speed and direction sensors and twelve air quality monitoring stations dot the Basin. In addition, during the summer of 1969, the Scott Research Laboratories (1970) carried out an extensive data gathering program in Los Angeles. Particularly valuable were the vertical temperature profile data they gathered over three sites in the Basin, thereby permitting much more accurate specification of the depth of the mixing layer than is normally possible. Finally, due to the severity and persistence of the smog which plagues the region, Los Angeles is the most appropriate area in which to apply such a model.

In this study, we applied Equations (8) to the prediction of pollutant concentrations over a fifty mile square area that includes virtually all centers of population in the Los Angeles Basin. This region, shown in Figure 1, was divided into a grid of 625 2 mile x 2 mile squares. Source emissions and meteorological variables are distributed according to this grid, i.e., two miles is the resolution of the model, or the spatial dimension over which all quantities are averaged. Furthermore, for reasons to be discussed, the grid actually used in the solution of Equations (8) is a three-dimensional array of ten layers of cells occupying the space between the ground and the base of the inversion and lying directly over the area shown in Figure 1. Thus, each cell has a base two miles square and a height of (H - h)/10. The center of each cell, or node, is the point to which values of all variables are assigned or referenced. Unfortunately, due to variations in both H and h with x and y and, in the case of H, with t, the three-dimensional modeling region has an irregular "roof" and "floor". To eliminate these irregularities, which hamper the solution of the equations, we performed the following change of variables

$$\xi = \frac{x}{x_E - x_W}$$

$$\eta = \frac{y}{y_N - y_S}$$

$$\rho = \frac{z - h(x, y)}{H(x, y, t) - h(x, y)}$$

thereby transforming Equations (8) to Equations (D-4), given in Appendix D. The entire airshed is thus transformed into a cube, one unit on a side, in the  $(\xi,\eta,\rho)$  space. The actual model is then based on the solution of Equations (D-4) over a dimensionless region comprised of 25 x 25 x 10 rectangular parallelopipeds.

The objective of this study, then, is to solve Equations (8), or equivalently, the transformed Equations (D-4), over a particular period of time, comparing the predicted concentrations with those measured at local air quality monitoring stations. A number of factors enter into the accomplishment of such a task:

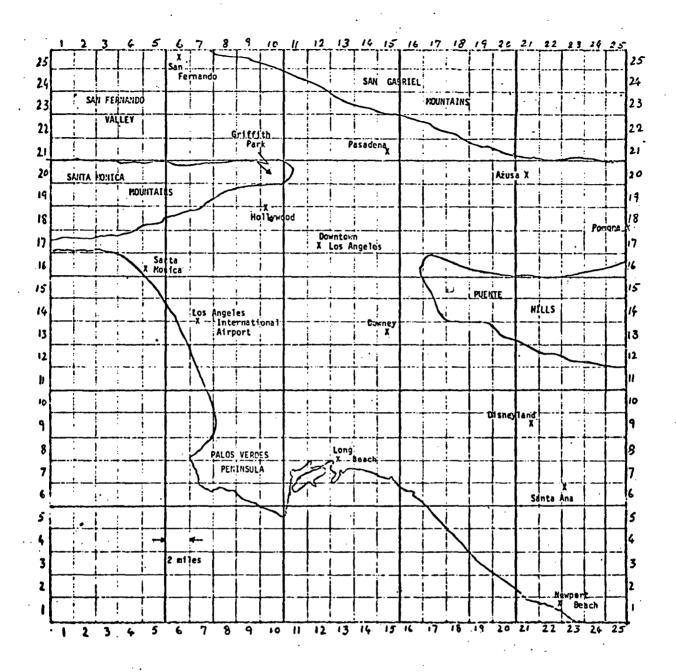


Figure 1. The Modeling Region

- The development of a kinetic mechanism capable of describing the rates of chemical reactions occurring in the atmosphere, and the adaptation of this mechanism for inclusion in the airshed simulation model.
- The selection and adaptation of a finite difference technique suitable for use in the numerical integration of Equations (D-4), the development of a computer program embodying the method, and subsequent testing of the method to determine its stability and accuracy characteristics.
- The development of a contaminant emissions inventory for the Los Angeles Basin.
- The development of a means for treating the various meteorological parameters, including the winds aloft.

Upon selection of particular validation days, in our case 29 and 30 September 1969:

- Maps must be prepared expressing the spatial and temporal distributions of surface wind speed and direction and the height of the inversion base.
- . Air quality data must be gathered.

Validation of the model is undertaken upon completion of these various efforts and is comprised of three parts, to be carried out in sequence.

- (1) Validation for carbon monoxide. The main purpose of this part is to provide a test of the meteorological facets of the model. If the model can be validated for CO, then confidence may be placed in the treatment of the winds and the inversion, and validation of the photochemistry may proceed. Validation for CO also constitutes a test of portions of the source emissions inventory and the numerical integration technique.
- (2) Sensitivity studies. Exploration of the effects on predicted CO concentrations of changes in meteorological variables, such as winds, inversion height, and diffusivities. Also, study of the effects of varying the order of the numerical technique, the size of the grid system (for example, one mile instead of two mile squares), the number of horizontal strata of cells, and the size of the integration time step.
- (3) Upon successful completion of (1) and (2), and upon incorporating modifications suggested by the results of the studies, carry out validation runs and sensitivity studies for hydrocarbons, nitrogen oxides, and ozone.

Clearly, the size and scope of such an undertaking is great. Since the inception of the project last July, we have fully completed the developmental effort, but we have undertaken only a small portion of the validation segment. In particular, we have carried out validation studies for CO for 29 September 1969 and have explored a few questions concerning sensitivity of the model. In section III, we report on our accomplishments to date in validation of the model. In the remainder of this section, we outline briefly what has been achieved in model development. The details of these various efforts are reported in six accompanying appendices.

- We have completed a comprehensive emissions inventory for the Los Angeles Basin. Particular emphasis was placed on the spatial and temporal distribution of emissions from motor vehicles, as vehicular emissions account for a preponderance of carbon monoxide, hydrocarbons, and nitrogen oxides in the Los Angeles atmosphere. Attention was also given to those sources which, while responsible for only a small proportion of emissions on an area-wide basis, contribute heavily to pollutant concentration levels in their own locale--airports, power plants, and refineries. See Appendix A, "Contaminant Emissions in the Los Angeles Basin--Their Sources, Rates, and Distribution", for a complete description of this effort.
- We have incorporated into the airshed model a new kinetic mechanism developed by Thomas A. Hecht and John H. Seinfeld of the California Institute of Technology. The results of validation studies demonstrate that this model is capable of predicting with acceptable accuracy the concentration/time behavior of smog chamber experiments for propylene, isobutylene, n-butane, and a mixture of propylene and n-butane at initial NO to hydrocarbon ratios of 1/3 to 1. The mechanism has also been shown to simulate accurately the effect on photo-oxidation rates of variations in CO concentrations, as well as the inhibitory effect of high initial concentrations of nitric oxide on the maximum concentration of ozone obtained. A full discussion of the mechanism and the validation results is given in Appendix B, "A Kinetic Mechanism for Atmospheric Photochemical Reactions."
- We have prepared hourly maps for each of the validation days that display spatial and temporal variations in surface wind speed and direction and in the height of the inversion base. We have also explored several approaches to the estimation of the wind field aloft, one of which appears to be a useful method for the automatic generation of these winds. We have investigated the use of computer graphics in the preparation and conversion to digital form of wind maps, and have attempted to develop a simple model to describe variations in mixing depth with time and location. A complete description of these efforts is presented in Appendix C, "The Treatment of Meteorological Variables."
- We have adapted the method of fractional steps for use in the finite difference solution of the equations of conservation of mass. Details of the method, and an evaluation of its stability and accuracy characteristics, are given in Appendix D, "Numerical Integration of the Continuity Equations."

- We have gathered together all the available air quality data for the validation days 29 and 30 September 1969. These data are presented in Appendix E, "Air Quality Data Used in Model Validation."
- . We have prepared a computer program to carry out the many calculations required in an airshed simulation. A general discussion of the program is presented in Appendix F, "Description of the Computer Program."

# III. VALIDATION OF THE MODEL

The major validation result obtained as of this writing is the prediction of the distribution of carbon monoxide concentrations over the Los Angeles Basin for the period 5 AM to 5 PM PST (6 AM to 6 PM PDT) on 29 September 1969. This validation run constituted a pivotal test of the treatment of meteorological variables, as well as of the accuracy of the emissions inventory and the suitability of the numerical method. (Actually, the integration of the coupled equations, with photochemical reaction terms included, is a much more severe test of the numerical technique.) The conditions under which the run was made are as follows:

- Meteorology Hourly maps of wind speed, wind direction, and mixing depth were prepared, as described (and examples of which are shown) in Appendix C, Sections I and II. Wind variables and inversion height were maintained constant throughout each hourly interval, commencing at the half hour. When the inversion base was raised, the concentration of carbon monoxide in the air newly included under the inversion was taken to be equal to the CO concentration at the inversion base just prior to its displacement upward. Vertical turbulent diffusivity was treated as shown in Appendix C, Section IV. The second method for constructing the wind field aloft, as described in Appendix C, Section III (p. 40, paragraph 2), was used.
- Emissions Carbon monoxide emissions from automobiles and aircraft (both on the ground and in flight) were included, as described in Appendix A. One modification, not discussed in Appendix A, was made in the treatment of vehicle emissions. To account for the higher emissions rates due to cold starts in the morning, we assumed that

$$Q_{CO} \left[ \frac{\text{grams}}{\text{vehicle mile}} \right] = \begin{cases} 63.9 \left( \frac{49-t}{40} \right) & 5 \le t \le 9 \\ 63.9 & 9 \le t \le 17 \end{cases}$$

where t = time (in hours PST)\*

- Numerical Integration The method described in Appendix D
  was used, applying the second order approximation in both the
  horizontal and vertical. The time step for the integration was
  two minutes.
- Validation data Predictions were compared with measured data,
   as reported in Appendix E.
- . Boundary Conditions

$$g_{CO}(x,y,z,t) = 3$$
 ppm at all boundaries

Initial Conditions - We used a 5 AM PST concentration map for carbon monoxide, as constructed from interpolation and extrapolation of data reported in Appendix E. This map is shown in Figure 3. Unfortunately, the data upon which the map was based are hourly averages and not

<sup>&</sup>quot;It is assumed that emissions at 5 AM are all "cold start," with the "cold start" rate being 10% higher than the "hot start" rate. Emissions at 9 AM and thereafter are assumed to be "hot start," and values between 5 AM and 9 AM are computed through linear interpolation.

instantaneous values of concentration. Thus, the average of the 4 AM to 5 AM and 5 AM to 6 AM hourly averages was used to estimate the instantaneous values at 5 AM.

The validation run was carried out on an IBM 360-67 computer. Twenty-nine minutes of computing time were required to simulate twelve hours, a ratio of 1:24. Contemplated modifications, as noted in Section IV, are expected to substantially reduce this ratio.

The results of the validation effort are displayed in Figures 4 through 10 and twelve computer printouts.

- Figure 4. A summary of comparisons between predicted and measured concentrations at eleven sites scattered throughout the Basin. (The locations of the monitoring stations, along with their proximity to major emissions sources, are shown in Figure 2.) Both the predicted and the measured concentrations represent hourly averages beginning at the hour given.\* Since a two-minute time step was used, each reported hourly prediction is the average of thirty calculated values.
- Figures 5 through 10. Plots of predicted and measured concentrations vs. time of day for each of the eleven monitoring stations. These plots convey the same information shown in Figure 4.\*\*
- computer Printouts. Twelve maps of predicted average hourly concentrations throughout the day (except for the maps representing the time periods 8AM to 8:20 AM and 8:20 AM to 9 AM). The indexing system (row and column numbers) shown on the printouts corresponds exactly to that used in Figures 1 and 2; thus, a predicted value of CO concentration can be located geographically, and its proximity to freeways, airports, and monitoring stations noted, by reference to these figures.

It is perhaps wise to precede the discussion of these results with a few comments concerning what should be expected in a comparison of predicted and measured carbon monoxide concentrations.

In any interpretation of results, it is important to note that a measurement, if presumed accurate, is representative of the CO concentration in only a small portion of the 2 mile x 2 mile grid square for which the concentration is predicted. The prediction represents the average concentration over the entire grid square; higher concentrations, as well as lower

<sup>\*</sup> All predicted values are reported as truncated integers (e.g., an average concentration of 5.85 is reported as 5, and not 6), all measured values as rounded integers. Rounding and truncation to integers adds a possible discrepancy of up to 1.5 ppm to that which already exists between predicted and measured values.

<sup>\*\*</sup>Note that in Figures 5 through 10 the lines connecting both experimental and predicted values of concentration are merely symbolic links. They do not represent interpolated values. Links are not drawn through experimental points of questionable accuracy, but these points are indicated on the maps. Furthermore, links are not drawn for periods in which experimental data are unavailable, as the resulting plots may be visually misleading.

concentrations, very likely exist in portions of the square. Thus, recognizing that line sources are not well represented by a grid system and that sensors are generally located near freeways or arterials (see Appendix E, Table E-2), predicted and measured values may be expected to differ.

Returning now to the results, and with the preceding comments in mind, it may be observed from Figures 5 through 10 that reasonable agreement has been achieved at a majority of the monitoring stations over the course of the twelve hour validation period. At certain stations, however, comparisons are poor—in some cases during the early hours, in others, over a period of several hours.\* These discrepancies may be categorized into two classes, each attributable to an identifiable deficiency in the model, in its formulation, or in the method of solution. Two of the deficiencies are correctable, and one is inherent to the model.

 Comparisons made at the Azusa (AZV) and El Monte (ELM) stations, both located in the San Gabriel Valley, are poor during the morning hours. This appears to be attributable to the means by which temporal variations in the height of the inversion base are treated in the model. The mixed layer is very shallow over the San Gabriel Valley early in the morning, increasing rapidly in depth through the morning hours. Recall, however, that the depth of the layer is altered only once an hour in the calculation. When the base of the inversion is displaced upward, the value of concentration at the base just prior to its vertical displacement is assigned to the entire volume of air newly included under the displaced inversion. While this hourly displacement occurs over all grid squares in the Basin. its effect on the calculation is most pronounced over the San Gabriel Valley, where the estimated mixing depth during the morning hours varies as follows:

Time (AM PST)	Mixing Depth (feet)
<b>05</b> 00 <b>-</b> 0530	60
<b>0530 -</b> 0630	60
<b>0630 - 0</b> 730	<b>150</b>
<b>0730 -</b> 0830	200
<b>0830 -</b> 0930	400
<b>0930 - 1</b> 030	650

Since the inversion is shallow, high concentrations build up rapidly at the top of the mixed layer, and when the inversion base is displaced upward, the equivalent of a large elevated source of CO is introduced. We believe that by altering the height

<sup>•</sup> The Pomona monitoring station lies one mile east of the eastern boundary of the modeling region. The concentration calculated at the closest grid square is compared with that measured at Pomona.

of the inversion base at each time step in the integration, (i.e., every two minutes), this effect can be alleviated.\*

- Predictions of peak CO concentrations, between 7 AM and 9 AM, are typically too low. Extreme examples occur at Lennox (LENX), West Los Angeles (WEST), and Whittier (WHTR); others occur at Commerce (VER) and Reseda (RESD). Since all monitoring sites, except for the station at Azusa, are located near major local sources, particularly freeways and arterials, the stations are probably "seeing" local high concentrations. The model, having a two-mile horizontal spatial resolution, will be incapable of predicting these local peak concentrations. This thesis may be explored further by applying the model to a smaller region, using a finer horizontal resolution.
- Perhaps equally significant is the effect on the accuracy of prediction of assuming that automotive emissions can be represented by a constant factor, Q<sub>CO</sub>. As we discussed in Appendix A (see pages A-1 to A-4, A-19 and A-26), the representativeness of the vehicular emissions factors depends on the extent to which the driving cycle upon which they are based actually simulates average vehicular emissions. Even if the cycle is adjudged to be representative, cognizance must be given to the fact that carbon monoxide emissions rates vary with percentage of time in an individual operating mode. In particular, increased contaminant emissions rates result from the increased frequency of acceleration, deceleration, and idle at the low average speeds that occur during periods of congestion. Thus, the CO emissions rate, taken to be constant in the validation run, does not properly represent the morning rush period. This deficiency in the treatment of emissions ray be alleviated by assuming that  $\mathcal{Q}_{CO}$  has a higher value during periods of congestion than during periods of normal traffic.

Discrepancies between prediction and measurement can be only partially rectified by correcting deficiencies in the model. The accuracy of prediction is dependent upon the quality and quantity of available meteorological and emissions data, as well as upon the "goodness" of the model. Furthermore, the magnitude of the discrepancies in the results depend, not only on the accuracy of the model's predictions, but also upon the accuracy of the air quality data. Thus, it would be inappropriate to reflect further on the results without commenting on the accuracy of the data.

The inversion maps, as was discussed in Appendix C, are based on temporal and spatial interpolation of vertical temperature profiles taken at about 8 AM and 1 PM at three points located between Los Angeles International Airport and El Monte. All values outside these ranges are extrapolated, and the accuracy of these extrapolated values is questionable. (The accuracy of the temperature profiles, themselves, is questionable, as altimeter readings sometimes registered 25 to 75 feet below ground elevation.) In addition, sensitivity studies have indicated that modest changes in the depth of

<sup>\*</sup> Since mixing depths over the San Fernando Valley are about the same as those over the San Gabriel Valley during the morning hours, similar discrepancies might be expected in the former location. These discrepancies are not observed; however, due to the existence of a diverging surface wind field in the San Fernando Valley. This divergence necessitates the supply of air from above, thereby reducing the concentrations at the base of the inversion and thus diminishing the magnitude of the elevated artificial source.

the mixed layer have a significant effect on predicted ground concentrations. It is therefore apparent that the number of sites at which vertical temperature soundings were made, and the number of soundings carried out daily at each site, are inadequate. In addition, the need for extrapolation in the early morning hours, and out to the borders of the modeling area (including the San Fernando Valley), should be minimized.

While the number of wind monitoring stations in the Basin is sufficient to give adequate definition to the surface wind field, the accuracy of the readings is often questionable. A substantial number of wind speed and direction measurements, taken at identical or adjacent sites at the same time, differ markedly. At El Monte, for example, Scott and LACAPCD made wind measurements at virtually the same site; their reported readings typically differ by 30% in speed and 30° to 45° in direction, and, on occasion, by 200% and 180°, respectively. Measurements made simultaneously at the Encino, Van Nuys, and Reseda stations in the San Fernando Valley often varied by as much as 180°, making the construction of wind maps in that area a dubious undertaking.

We turn at this point to the computer printouts, representations of predicted values of average hourly concentrations of carbon monoxide over the entire 50 mile x 50 mile grid. (Note that the period 8 AM to 9 AM is represented by two maps--8:00-8:20 and 8:20 - 9:00.) The reader should refer to Figures 1 and 2 to establish the correspondence between predicted concentrations and geography. In general, the predicted values shown on the printouts appear to give a reasonable representation of temporal variations in the concentration field. The morning buildup of carbon monoxide is clearly demonstrated, followed by a mid-day cleansing of the Basin as the winds blow the pollutants northward and out of the modeling area. Low afternoon concentrations then generally prevail.

Large concentration gradients and apparently random fluctuations may be noted at certain locations on virtually all the maps. When these occur in the vicinity of a monitoring station, an average value of concentration, weighted according to distance from the four closest node points, is reported in Figure 4 (and thus in Figures 5 to 10). For example, ELM, located in the northwest quadrant of ground square (18, 18), has a predicted average hourly CO concentration of 19 ppm between 9 AM and 10 AM PST. The average hourly concentrations in the local area, as may be seen from the computer printout, and as shown here,

14	16
17	X ELM 21

vary greatly. Thus, the predicted value of concentration in areas of large gradients has a relatively large uncertainty associated with it. This should be kept in mind when evaluating the results of Figures 5 to 10.

Two problems arise in the northwestern portion of the Basin, which includes the San Fernando Valley, that become apparent upon viewing these maps. The first is that the difficulties encountered in representing the

wind field in this area manifest themselves in peculiar ways. Very often large vertical components of the wind are required in order to satisfy continuity, and, as these appear to be distributed somewhat randomly throughout the Valley, the resulting concentration field is more a series of fluctuations than the smooth field characteristic of the rest of the modeling region. Second, anomalies sometimes occur in regions in which horizontal spatial gradients in emissions rates are steep. For example, row 21 in the northwest includes a large emissions source, the Ventura Freeway, while row 20 lies along the Santa Konica Mountains, a region in which virtually no emissions occur. The finite difference method, in approximating the large concentration gradients that exist in this region, often predicts depressed and, on occasion, negative values of concentration. The use of higher order approximations may be needed to eliminate this problem.

To summarize, then, we believe that reasonable agreement has been achieved at a majority of monitoring stations. Where discrepancies between prediction and measurement are great, assignable causes can be identified. Some are correctable, others are inherent in the nature of the approach and in the spatial resolution of the model. Finally, inaccuracies in source emissions and meteorological data and in their representation contribute significantly to the discrepancies that are observed.

The remaining validation efforts of interest can be described very briefly. We carried out a validation run for 30 September 1969 similar to that of 29 September--prediction of carbon monoxide concentrations from 5 AM to 1 PM. Comparisons between prediction and measurement were less satisfactory than those obtained for 29 September. We believe that the relatively large discrepancies observed are attributable to poor definition of the wind field in the morning. Wind speeds were consistently 1 to 2 miles per hour over much of the Basin from 5 AM to 8 AM, a speed that is below the reported threshold of the measurement apparatus. We plan to examine the actual meteorological data in detail before proceeding further with validation runs on the 30th. (It should be noted, however, that the high concentrations experienced on that day make it most interesting for study.) We have not yet undertaken validation of the full photochemical model (including hydrocarbons, nitrogen oxides, and ozone) for either of the validation days as of this writing, although tests of the numerical method, as applied to the coupled equations, have been carried out.

A number of sensitivity runs were performed during the validation process, and the following observations were made. Modest variations in specification of the wind field and the inversion have significant effects on the magnitude and distribution of predicted concentrations. Sensitivity of the calculation to changes in the vertical diffusivity profile have not been investigated; however, vertical gradients are generally small, and predictions should not be greatly influenced by modest changes in the profile. Recent tests of the finite difference method, comparing the accuracy of the second and fourth order approximations in the horizontal when a second order approximation is applied in the vertical, indicate that only minor differences in predicted values result when the higher order method is used. However, for the validation run reported here, large concentration gradients are present and the two approximations may give somewhat different results in the immediate locale of these gradients. Finally, variations in

Q<sub>CO</sub> as a function of "degree of congestion" has not been investigated. However, it is expected that inclusion of a variable emissions rate will result in more realistic predictions of morning CO peaks.

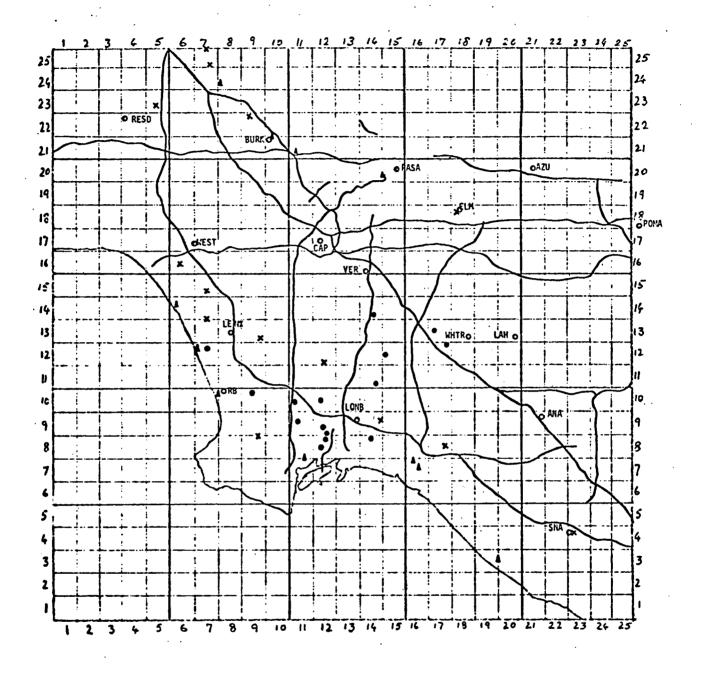


Figure 2. Locations of Monitoring Stations Relative to Major Contaminant Sources in the Los Angeles Basin

- freeways
- - oil refineries
- ▲ power plants
- = airports
- contaminant menitoring stations

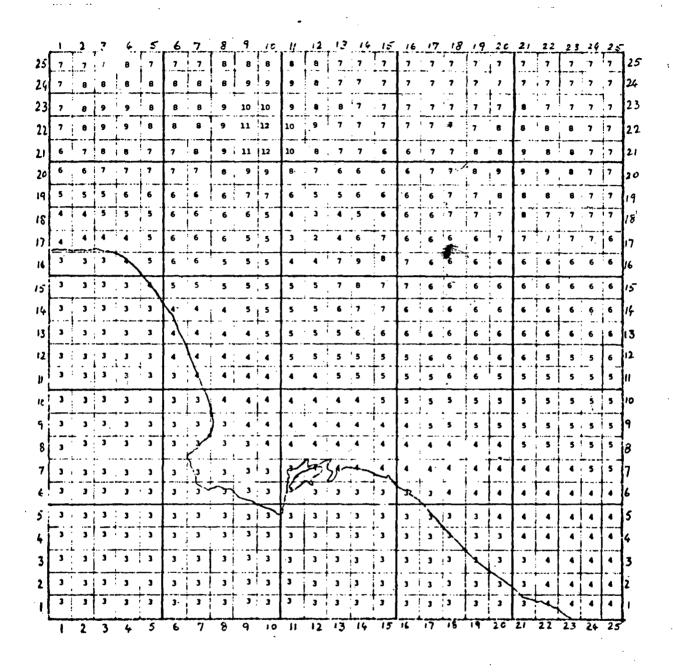


Figure 3. Initial Conditions - Carbon Monoxide Concentrations for 29 September 1969, 500 PST (in ppm)

Figure 4. Summary of Validation Results for Carbon Monoxide for 29 September 1969 (in ppm)\*

Sta	tion/Time (PST)	5 6	6 - 7	7 - 8	8 - 9	9 - 10	10 - 11	11 - 12	12 - 13	13 - 14	14 - 15	15 - 16	16 - 17
1	CAP	3 (5)	7 (12)	17 (17)	18 (20)	- (19)	- (13)	6 (7)	3 (4)	3 (4)	2 (4)	6 (5)	6 (6)
60	AZU	10 (9)	10 (11)	10 (13)	11 (13)	10 (14)	8 (16)	6 (15)	7 (3)	9 (6)	6 (4)	6 (4)	4 (5)
69	BURK	13 (8)	15 (11)	18 (14)	17 (14)	9 (13)	11 (12)	10 (9)	11 (9)	7 (4)	7 (3)	5 (3)	5 (4)
71	WEST	5 (6)	9 (9)	17 (10)	17 (10)	8 (8)	- (8)	5 (7)	4 (5)	4 (4)	4 (4)	4 (5)	5 (5)
72	LONB	7 (4)	9 (5)	13 (7)	11 (7)	9 (4)	6 (4)	6 (4)	6 (4)	5 (4)	5 (4)	5 (4)	5 (4)
74	RESD	10 (9)	12 (10)	16 (12)	11 (10)	8 (10)	6 (11)	4 (8)	5 (4)	3 (3)	3 (4)	5 (4)	5 (4)
75	POMA	7 (6)	8 (6)	9 (5)	9 (5)	<b>-</b> (5)	6 (5)	5 (8)	4 (9)	5 (7)	6 (6)	6 (4)	6 (3)
76	LENX	6 (5)	15 (5)	9 (6)	5 (5)	5 (4)	6 (4)	- (4)	3 (3)	3 (3)	4 (3)	6 (3)	6 (3)
80	WHTR	- (6)	11 (5)	14 (7)	11 (11)	11 (13)	13 (13)	- (10)	. 2 (2)	1 (4)	1 (4)	1 (4)	1 (4)
(sc	:) ELM	- (8)	- (11)	- (16)	8 (17)	6 (19)	6 (14)	9 (8)	5 (6)	3 (3)	2 (2)	2 (3)	- (3)
(sc	C) VER	9 (8)	12 (8)	17 (11)	15 (13)	15 (11)	9 (8)	6 (7)	4 (5)	3 (5)	3 (4)	4 (4)	3 (5)

<sup>\*</sup>The left-hand figure in each square is the measured value; the figure in parenthesis is the predicted value. All values are in parts per million of carbon monoxide, averaged over a period of one hour.

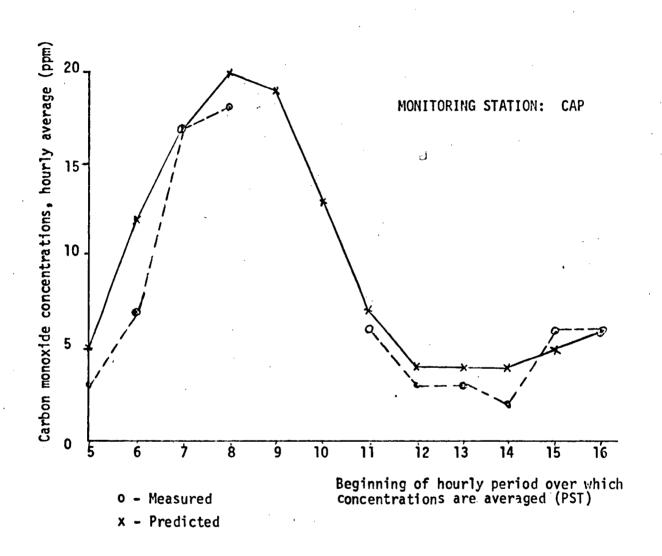
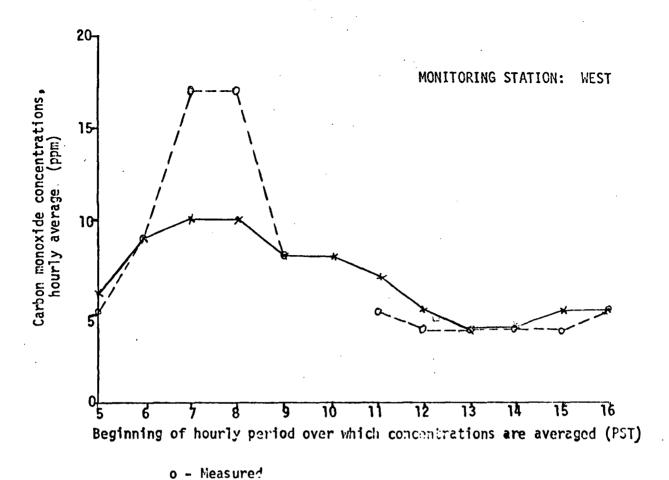


Figure 5. Temporal Variations in Predicted and Measured Carbon Monoxide Concentrations for 29 September 1969: Downtown Los Angeles



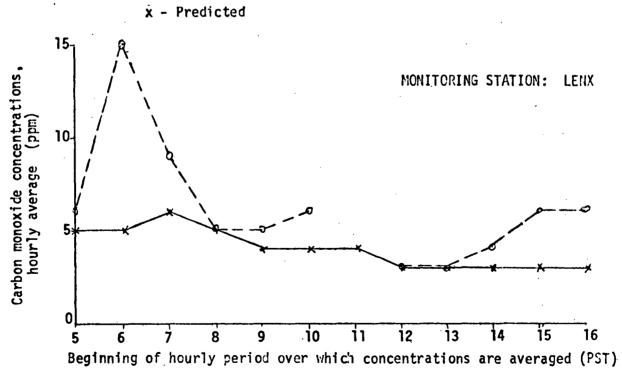
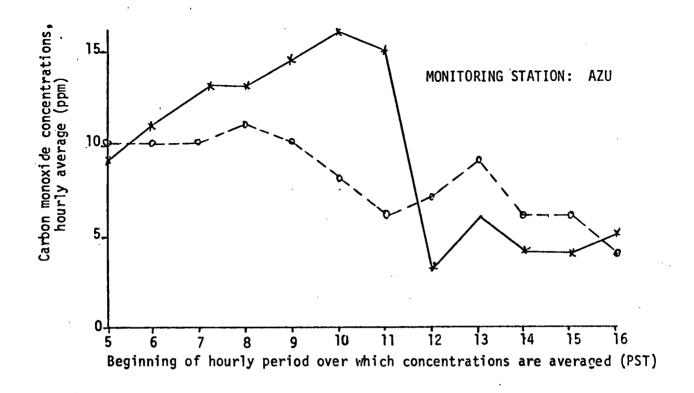


Figure 6. Temporal Variations in Predicted and Measured Carbon Monoxide Concentrations for 29 September 1969: West Los Angeles and Lennox



o - Measured
x - Predicted

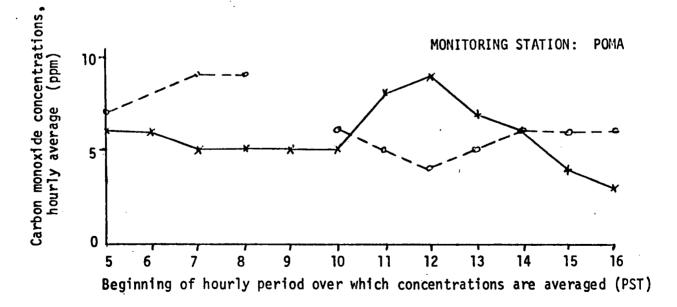


Figure 7. Temporal Variations in Predicted and Measured Carbon Monoxide Concentrations for 29 September 1969: Azusa and Pomona

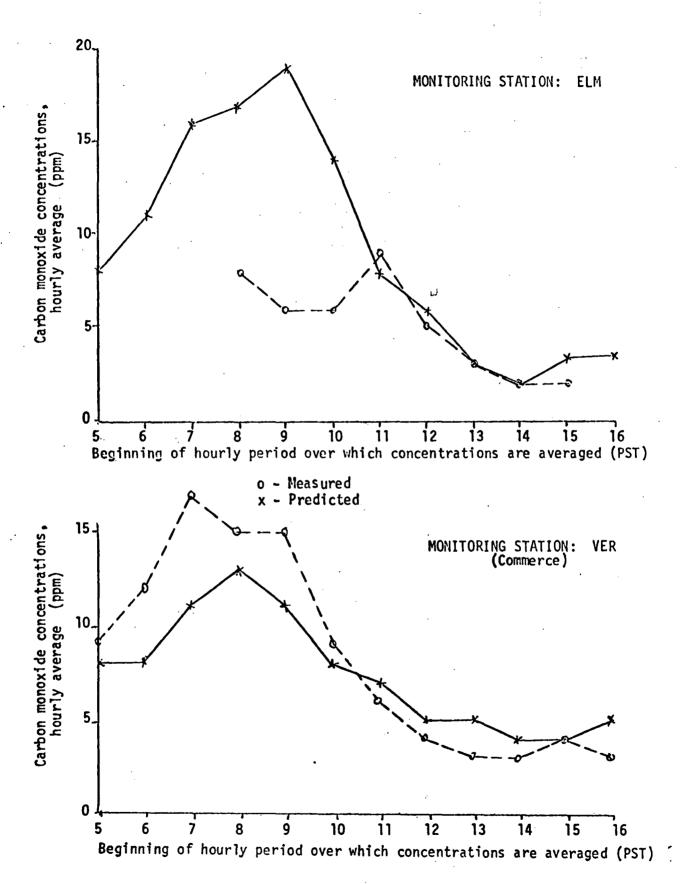
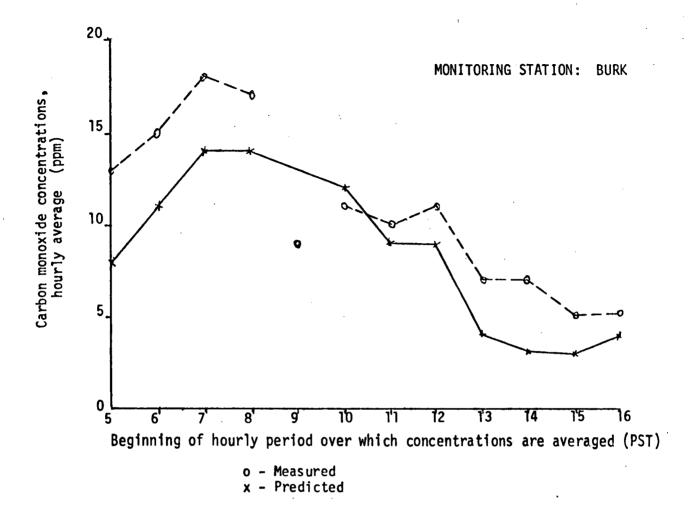


Figure 8. Temporal Variations in Predicted and Measured Carbon Monexide Concentrations for 29 September 1969: El Monte and Commerce



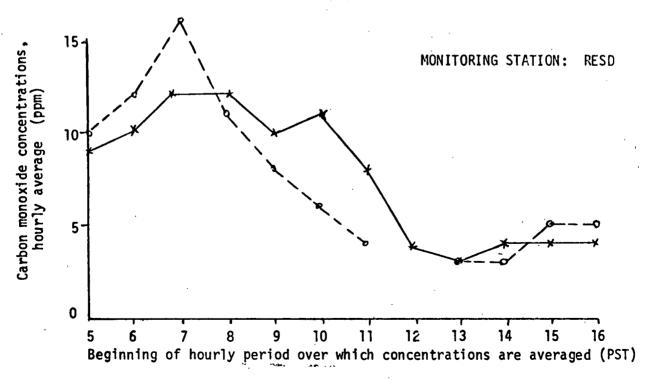
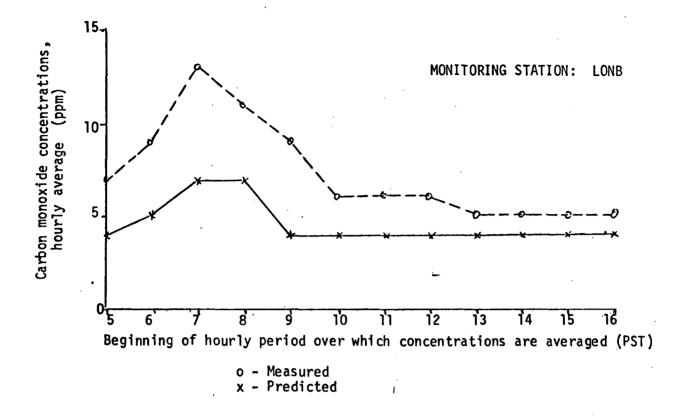


Figure 9. Temporal Variations in Predicted and Measured Carbon Monoxide Concentrations for 29 September 1969: Burbank and Reseda



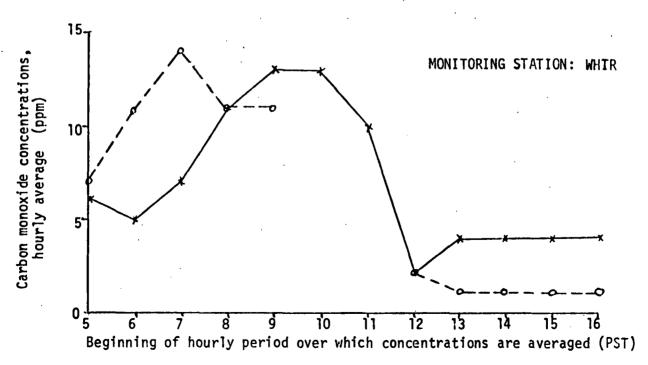


Figure 10. Temporal Variations in Predicted and Measured Carbon Monoxide Concentrations for 29 September 1969: Long Beach and Whittier

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#### IV. RECOMMENDATIONS

The accomplishments of the past year have been described in detail in earlier sections and in the Appendices. These include the formulation and development of an airshed model, the acquisition and preparation of emissions inventories and meteorological data, and the completion of the initial phase in model validation. The potential of the model for accurate prediction of the concentration distribution of inert species such as carbon monoxide may be assessed from the results described in Section III. But much remains to be accomplished. In this section, we present our recommendations for future efforts.

The recommendations which follow are divided into two categories, those to be undertaken in the near term and those to be initiated upon validation of the model for photochemical pollutants. (These latter recommendations will be referred to as "intermediate term".) The near-term efforts consist of the correction of model deficiencies which were noted in the discussion of the validation results, and validation of the model for photochemical pollutants. Also, we would investigate the reliability of the meteorological data base—in particular, wind measurement in near-calm conditions—and ascertain the effect on prediction of the probable imprecision of measurements made under these conditions. Upon successful validation of the model we would begin the intermediate—term efforts. These include the automation of meteorological calculations, the initiation of extensive validation studies and the optimization of the computer program to reduce computing time and computer storage requirements. More specifically, the tasks envisaged are as follows:

### Near Term

- A. Exploration and correction of model deficiencies revealed to date.
  - Incorporate means to account for high vehicle emissions rates occurring under conditions of heavy traffic congestion, especially on freeways.
  - Explore the effect on accuracy of prediction of certain aspects of the numerical technique. In particular, these include the order of integration method, the size of the surface grid, and the number of horizontal strata employed.
  - 3. Investigate sources of error and degree of inaccuracy of wind measurements made at low wind speed, especially in relationship to the early morning September 30 data.
  - 4. Evaluate the sensitivity of calculation to the form of the diffusivity relationship.
- B. Validation of Model for Photochemical Pollutants. In addition to the usual aspects of validation, this effort will include:

- Adaptation of the kinetic mechanism to describe the atmospheric reaction mixture (thus far the mechanism has only been applied to smog chamber data, as described in Appendix B). Also, inclusion of the effects on reaction rates of the changing composition of the atmospheric hydrocarbon mixture and the presence of carbon monoxide and water. These efforts are directed toward establishing guidelines for selecting generalized rate constants and stoichiometric coefficients.
- 2. Determination of the best method for representing the atmospheric hydrocarbon mixture. This effort will focus on the choice of the number of groupings of hydrocarbons that are to be employed, with consideration being given to both accuracy of prediction and computing time.

# Intermediate Term

# A. Automation of Meteorological Calculation

- Develop a new method, or modify an existing technique, for the automatic calculation of wind speed and direction in each cell using meteorological data obtained at ground stations scattered throughout the Basin. Such a method, most likely based on the interpolation of ground data, must be suitably adapted to Los Angeles' unique topography.
- 2. Codification and extension of Edinger's model of inversion behavior to provide for automatic calculation of mixing depth.

### B. Model Improvements

- 1. Examine alternative computational methods to determine whether computing time can be shortened without loss in accuracy, and whether accuracy can be improved without an increase in computing time.
- 2. Redefine the modeling area to exclude many of the cells lying over the ocean.
- 3. Investigate the possibility of carrying out a simulation throughout the night and into the next day. A major problem associated with such a calculation is the lack of vertical temperature profile data at night; however, it may be possible to consider just one shallow well-mixed layer for this period.
- 4. Optimize the current computer program to reduce computing time and computer storage requirements.

### C. Sensitivity and Validation Studies

- 1. Investigate sensitivity of predicted concentrations to variations in model parameters and boundary conditions.
- 2. Carry out extensive validation studies for differing types of meteorological conditions.

It should be kept in mind that as progress is made and improvements in prediction are realized, an ever greater proportion of the discrepancy between predicted and measured values of pollutant concentrations will be attributable to the limitations in quantity, representativeness, and accuracy of meteorological, source emissions, and air quality data. Thus, in addition to the refinements recommended above, attention must be given to the acquisition and incorporation of a richer and more accurate data base as efforts in modeling proceed.

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