MODELING ATMOSPHERIC EFFECTS - AN ASSESSMENT OF THE PROBLEMS

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ABSTRACT

Our ability to simulate atmospheric processes that affect the life cycle of pollution is reviewed. The transport process is considered on three scales (a) the near-source or single-plume dispersion problem, (b) the multiple-source dispersion problem, and (c) the long-range transport. Modeling the first of these is shown to be well within the capability of generally available techniques, although determining the input parameters is often difficult. The second scale has been well studied within the context of urban diffusion and is a very productive area of current research. Finally, long-range transport is treated mainly as a meteorological problem. The state-of-the-art in modeling the various meteorological processes is reviewed. Removal of pollutants is discussed in the form of both dry deposition and precipitation scavenging. It is suggested that dry removal is especially effective in forested areas, and that the forests may enhance accumulation of pollutants.

An approximate transport model is developed which is used to calculate the ambient concentration of SO_2 throughout the United States. Associated calculations include dry deposition and the ratio of dry to wet removal for each State of the contiguous United States.

1. INTRODUCTION

It is generally accepted that precipitation in the Scandinavian countries may have values of pH sufficiently low to cause damage to terrestrial and aquatic ecosystems. The low pH is attributed to anthropogenic sources of SO_2. The location of these sources is somewhat open to discussion, although it is clear that Eastern and Western Europe and England are all contributors. The acidity of precipitation in the northeastern United States appears to be increasing.

A particularly insidious point about acid precipitation is the fact that the sources are not necessarily close to the most acid measurements. The U.S. has set maximum standards of groundlevel concentration of SO_2. A common method for achieving these standards is to release the SO_2 through tall stacks so that it will be dispersed and reduced to an acceptable level of concentration prior to reaching the ground. This has been a rather short-sighted solution, however, since it in no way reduces the amount of SO_2 emitted into the air.

In order to control increasing levels of SO_2 in the air, the U.S. Environmental Protection Agency has promulgated new source performance standards. These laws require that no more than a small amount of SO_2 may be emitted from any new sources. Nevertheless, there are so many new SO_2 sources proposed and under construction that these regulations will certainly be subject to a thorough review.

This paper will attempt to provide some background on how to relate source emissions to ambient concentrations. It will review some of the methodology for developing air quality models, present some of the existing models, and consider needed improvements. Finally, to provide some background to the conference, I have attempted to put the U.S. problem of SO_2 concentrations in perspective with some approximate calculations.

A major objective of this analysis is to point up the need to study the long-range transport of pollution. Dry and wet removal mechanisms as outlined in this paper suggest that forested areas may be a major recipient of air pollutants removed from the atmosphere. Thus the problem of acid precipitation, indeed the problems of long-range transport and of background air quality, may eventually affect the productivity and multiple uses of our forests.

In the next section, the general theory of air pollution modeling is reviewed. Section 3 deals with the single source and the methods developed for calculating dispersion from such a source. Section 4 on multiple source needs is largely a state-of-the-art review on urban modeling, since this represents the bulk of the work on this scale. Section 5 deals less with the methodology of evaluating long-range transport and more with the complexity of mesoscale meteorology. In Section 6, the dry deposition of air pollutants, particularly SO_2, is reviewed in depth, with some general discussion of wet removal processes. The paper concludes with an assessment of U.S. ambient SO_2 levels in Section 7.

2. THE GENERAL TRANSPORT PROBLEM

The behavior of the atmosphere determines in large part the transport of materials introduced into the air. Three elements of the

atmosphere which affect its behavior are the wind field, the temperature structure, and the various forms of the water substance. In dealing with problems of acid precipitation all three of these elements play a major contributing role. Wind fields exhibit a wide range of scales containing significant amounts of energy. The temperature field, in response to solar forcing, determines in large part the degree of mixing within an air mass. The occurrence and nature of the precipitation affect the cleaning of the atmosphere.

The problem of transport can be reduced to solving a complex equation representing the conservation of mass. Written in terms of the concentration of a particular chemical species, say C_α, this becomes

$$\frac{\partial}{\partial t} C_\alpha + \underset{\sim}{V} \cdot \underset{\sim}{\nabla} C_\alpha = S_\alpha - R_\alpha + \kappa_\alpha \nabla^2 C_\alpha \qquad (1)$$

where

$\underset{\sim}{V} = u, v, w$ = velocity vector

$\nabla = \frac{\partial}{\partial x}, \frac{\partial}{\partial y}, \frac{\partial}{\partial z}$ = space differentiating operator

S_α = sources of species α

R_α = sink of species α

κ_α = molecular diffusivity of species α

It should be noted that the precise specification of $\underset{\sim}{V}$, S_α and R_α is a rather complex problem. Generally these terms are dependent on time, space, chemical reaction, and surface conditions, among other factors.

Physical transport is complicated by the fact that the atmospheric velocity field, separated into components (u, v, w) in each of three coordinate directions, is not a constant in either time or space. Velocity measured at a point varies with a very high frequency, indicating that significant motion changes occur on time scales shorter than one second. High frequency variations must be filtered from any trends of a long-term nature in order to access the effect of the velocity field on transport. This is done by introducing an average or filtering assumption, by which all the variables are redefined as a mean value $\bar{A}$ and a fluctuating component A'

$$A = \bar{A} + A'$$

The equation is then averaged using the mean value definition to give the following:

$$\frac{\partial}{\partial t} \bar{C}_\alpha + \underset{\sim}{\bar{V}} \cdot \underset{\sim}{\nabla} \bar{C}_\alpha = \bar{S}_\alpha - \bar{R}_\alpha + \kappa_\alpha \nabla^2 \bar{C}_\alpha - \underset{\sim}{\nabla} \cdot \overline{C'_\alpha \underset{\sim}{V'}} \qquad (2)$$

The equation is different because of the addition of a so-called "turbulent correlation" term. This changes the solvable problem of one equation for one unknown into an insolvable problem of one equation for two unknowns ($\bar{C}_\alpha$, $\overline{C'_\alpha \underset{\sim}{V}'}$). Additional information must be introduced in order to solve the equation. Generally, the turbulent correlation term is interpreted as a flux of species α across some surface due to the turbulent velocity $\underset{\sim}{V}'_\alpha$. As such, it is assumed that

$$\underset{\sim}{\nabla} \cdot \overline{C'_\alpha \underset{\sim}{V}'_\alpha} = - \underset{\sim}{\nabla} \cdot K_\alpha \, \underset{\sim}{\nabla} \bar{C}_\alpha \tag{3}$$

which formally defines K_α the eddy diffusivity of the α species. It is easily shown (Pasquill, 1962) that the eddy diffusivity K_α is much larger than the molecular diffusivity κ_α and that the latter term can be neglected in equation (2). Thus we result with

$$\frac{\partial}{\partial t} \bar{C}_\alpha + \underset{\sim}{\bar{V}} \cdot \underset{\sim}{\nabla} \bar{C}_\alpha = \bar{S}_\alpha - \bar{R}_\alpha + \underset{\sim}{\nabla} \cdot K_\alpha \, \underset{\sim}{\nabla} \bar{C}_\alpha \tag{4}$$

as a representation of the conservation of mass.

Significance has been attached to the difference between the second and the last terms in equation (4). The second term represents advection or bulk movement of the average concentration by the average velocity. The last term represents the diffusion of material by the turbulent velocity field. If we consider a spherical cluster of particles released in the atmosphere, for example, the advection term would transport these particles from their release point according to a trajectory determined by the mean velocity, while the diffusion term would spread the particles by mixing them with the atmosphere. Most considerations in atmospheric transport are based upon a simplification and idealization of either or both of these processes.

In general, the simplification which is most appropriate depends upon the scale of the mixing process under consideration. We will identify three scales and concentrate on them. The near-source diffusion problem is rather classically treated by neglecting advection. Some problems related to developing the parameters necessary to use the models are discussed in Section 3. The multiple-source diffusion problem deals with the mixing of individual plumes into each other. This problem has been treated by a wide variety of techniques with little consensus on any one proper approach. The details of the wind field variations in both time and space must be considered with regard to their mixing properties, but this may be accomodated in different ways as described in Section 4.

Perhaps the most difficult scale to deal with is the more regional scale ranging into thousands of kilometers. The methods for modeling on these scales are only just beginning to be developed.

While we will concern ourselves with the diffusion and advection terms of equation (4), the values of the source and sink terms S_α, R_α,

warrant discussion as well. The source term represents the input of α to the total amount of α, and the sink term represents the removal of α. These terms are complex because of both atmospheric reactions and physical removal mechanisms. The latter will be discussed in Section 6, but the former are beyond the scope of this paper. If, for example, α is sulfur dioxide, then the source term should consist of the emission of SO_2 from both point and area sources as well as atmospheric chemical processes, such as oxidation of H_2S, that produce SO_2. Chemical processes, also deplete SO_2 by various oxidation reactions requiring their consideration in the sink term.

The most common procedure for dealing with reactions is through chemical kinetics expressions for the rate of formation of a compound. Such equations in terms of concentrations may take the form

$$\frac{\partial}{\partial t} C_\alpha = - kC_\alpha C_\beta \qquad (5)$$

where k is a rate constant for the reaction

$$\alpha + \beta \rightarrow \gamma + \delta$$

Similar expressions can be written for C_β, C_γ, C_δ, so that all of the reaction components can be accounted for. Equation (5) represents a sink term for α and β and a source term for γ and δ~.

Arguments related to turbulence apply to equation (5) so that this equation can be rewritten as:

$$\frac{\partial}{\partial t} \bar{C}_\alpha = - k\bar{C}_\alpha \bar{C}_\beta - k\overline{C'_\alpha C'_\beta} \qquad (6)$$

which then introduces a coupling between the fluctuation of concentration (resulting from turbulence) and the fluid motion. From these equations it is obvious that the separation of transport phenomena from transformation phenomena is not strictly valid.

To understand the details of atmospheric transformations it is necessary to use the complete system of equations. Recently, Donaldson and Hilst (1972) have discussed the limitations of not considering turbulent correlations in equation (6). Their results illustrate two basic regimes; (a) where the transformation is fast and thus limited by the availability of reaction components, i.e., diffusion limited, and (b) where the chemistry is relatively slow and the turbulent correlation $\overline{C'_\alpha C'_\beta}$ becomes increasingly significant, i.e., mixedness limited. These added turbulence terms may be significant when dealing with relatively slow reactions in regions of high concentration although further information is needed on this point.

3. NEAR-SOURCE DIFFUSION

Consideration will be limited to diffusion of materials emitted from an elevated point source, since the vast majority of SO_2 emitted into the atmosphere is through this mechanism. The concentration of SO_2 emitted from such a source, while quite variable, is of the order of 10^9 $\mu g/m^3$. Generally, the emission is hotter than the surrounding air, and is moved away from the stack by both buoyancy forces and by momentum imparted by the exit velocity. The maximum height to which the plume rises depends upon a set of parameters including the exit velocity, exit diameter, temperature difference, and the atmospheric temperature profile.

The emission concentration can easily be reduced two or more orders of magnitude within the plume a short distance from the source as the plume spreads due to mixing with air. It has been both empirically observed and theoretically derived that, under certain ideal conditions, the spread of material in the plume will exhibit a gaussian shape. This distribution, for example, is a valid solution to equation (1) if one reduces it to:

$$\frac{\partial}{\partial t} C_\alpha = K_\alpha \left(\frac{\partial^2 C_\alpha}{\partial x^2} + \frac{\partial^2 C_\alpha}{\partial y^2} + \frac{\partial^2 C_\alpha}{\partial z^2}\right)$$

and considers the K a constant related to a spreading parameter of the gaussian plume. Sources of more detailed information include Slade (1968), Turner (1970) and Pasquill (1962) which provide a discussion on the validity of the gaussian model and further rationale for its use.

The generally accepted formula used to calculate diffusion from a point source is

$$\chi = \frac{Q}{\pi \sigma_y \sigma_z \bar{u}} \exp[-y^2/2\sigma_y^2 - h^2/2\sigma_z^2] \qquad (7)$$

where

χ = concentration, Q = emission rate

σ_y = standard deviation of the gaussian plume in the y horizontal coordinate direction transverse to the wind direction.

σ_z = standard deviation of the gaussian plume in the z vertical coordinate direction

$\bar{u}$ = mean wind speed in the x direction

h = effective stack height = $h_s + \Delta h$ = stack height + plume rise

The plume is assumed to be oriented with the mean wind, which for the duration of the calculation is assumed to be steady (not changing in direction) and constant (not changing in magnitude). Determination of the diffusion parameters σ_y and σ_z is by far the most difficult task in the computation. The difficulty is relating the values of σ_y and σ_z to other more conventional and easily measured meteorological parameters. The parameters are strong functions of turbulence in the atmospheric boundary layer. A number of methods are available to establish numerical relationships, largely based upon experimental studies performed in England and the United States. The most conventional systems include about six separate categories of atmospheric stability -- so called classes A, B, C, D, E, and F -- ranging from the most unstable and most highly turbulent (A) down to the most stable (F). For example, Briggs and Gifford at Oak Ridge National Laboratory (Gifford, 1974) have recently analyzed the various diffusion parameterizations and suggest that, for class A stability

$$\sigma_z = 0.2\bar{X}$$

where $\bar{X}$ is the horizontal distance from the source, while for the most stable class F

$$\sigma_z = 0.02\bar{X}/(1 + .0003\bar{X})$$

or roughly an order of magnitude decrease in the dispersion parameters. Curves of σ_y and σ_z as a function of stability class are presented by Turner (1970), and appear as figures 1a and 1b.

The next difficulty is relating measured atmospheric conditions to the various stability classes. Atmospheric turbulence is generated both mechanically and thermally. These two types of forces are governed by different parameters, and the resulting turbulence is at most times a combination of the two. The roughness, Z_o, and the friction velocity, $U_* = -\overline{u'w'}$, appear as parameters of the wind profile given by

$$\bar{u} = \frac{U_*}{k} \ln(Z/Z_o) \qquad (8)$$

where k=.4. These are the mechanical roughness parameters. Thermal turbulence is governed by the incoming solar radiation and surface characteristics, but is measured by such parameters as the amount of cloud cover and the temperature change with height. A convenient parameter is the Monin-Obukov length scale, L,

$$L = -\frac{U_*^3 \, T}{kg \, \overline{w'T'}}$$

(where T = reference temperature, g = acceleration of gravity and $\overline{w'T'}$ = vertical flux of heat) which represents the ratio of mechanical to thermal turbulence. The absolute value of this length scale is

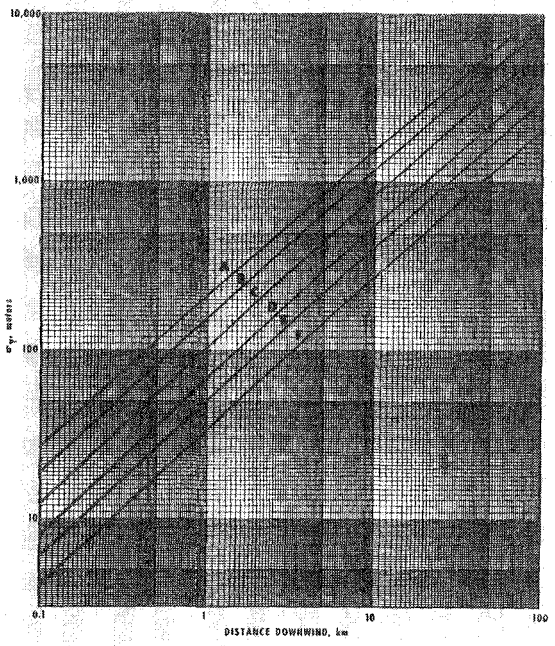

Figure 1a. Horizontal dispersion coefficient, σ_y, as a function of downwind distance from the source (Turner, 1970).

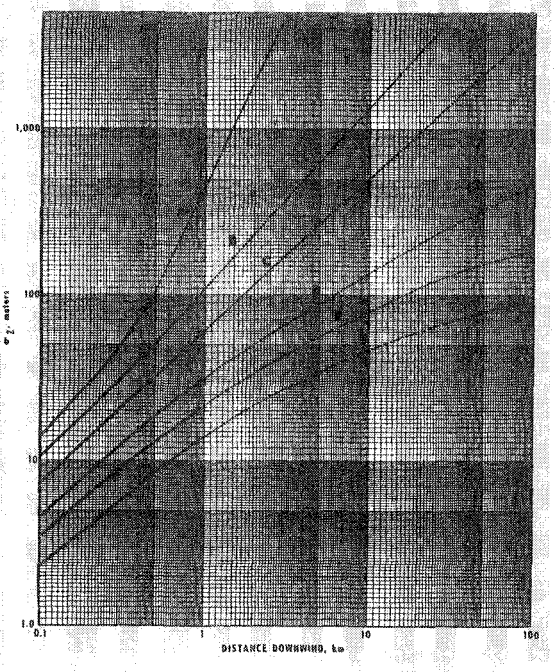

Figure 1b. Vertical dispersion coefficient, σ_z, as a function of downwind distance from the source (Turner, 1970).

proportional to the depth through which thermal turbulence is not significant. When $\overline{w'T'}$ is negative (heat is transported by turbulence toward the ground) L is positive and approaches infinity as the value of heat flux approaches zero. Smith, (1974) has recently related the various meteorological parameters namely incoming solar radiation, Z_o, U_*, $\bar{u}$ (10 meters) to the stability class. Figure 2 illustrates the relationships which result.

The remaining parameter in equation (7) is the plume rise Δh. Plume rise has been a subject of discussion for many years in the literature. The summary by Briggs (1969) is the generally accepted reference, although new data are appearing regularly. The formula which seems most appropriate for us here, valid for tall buoyant point sources, can be written as

$$\Delta h = \frac{1.6\ F^{1/3}\ x^{2/3}}{\bar{u}}$$

where

$$F = \frac{gQ_H}{\rho C_p \pi T_A}, \quad Q_H = \text{heat emission}, \quad T_A = \text{ambient temperature}$$

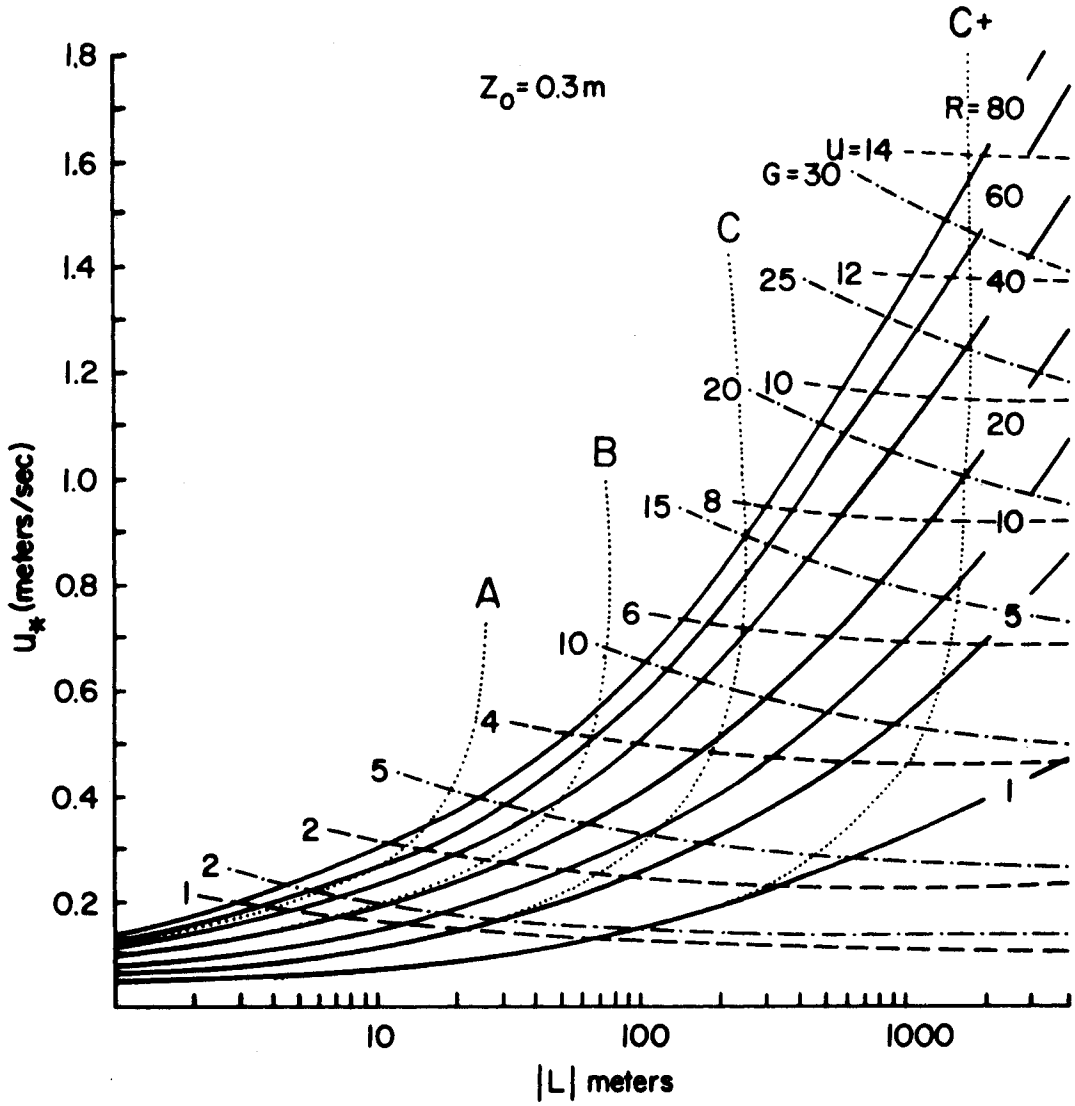

Figure 2. Relationship between solar radiation, R, Geostrophic gradient, G, wind at 10 meters, U, friction velocity, U_*, Monin-Obokov, length L and Pasquill stability classes, A-C, from a model of the boundary layer by F. B. Smith (1974).

The problem of estimating near-source diffusion has thus been reduced to relating meteorological parameters to source emission using various formulas presented here and other reference material. It is necessary to have meteorological information available, preferably from on-site measurements, in order to estimate the resultant diffusion.

4. MULTIPLE-SOURCE DIFFUSION

Multiple-source diffusion is generally studied in the context of an urban area. The approaches used range from distributing the emission uniformly through a volume defined by either arbitrary or natural boundaries, a so called box model, to a rather detailed solution of equation (2).

One logical method for calculating the diffusion from multiple sources is to advance the concept stated in equation (7). With a single direction strong wind persisting over the area, the summation of isolated point sources is mathematically straight forward and is a reasonable solution to the problem. When the wind is less pronounced, and the more variable, models have been developed which employ a gaussian distribution for a discrete set of sources. Such models have been discussed extensively in the Proceedings of the conference on urban diffusion models (Stern, 1970).

A simple material balance over a volume defined by cross sectional area, A(H) where H is the depth of the inversion which limits the area and h is the length would yield a concentration given by

$$\chi = Q/A(H)L \tag{9}$$

if there is no "ventilation". If, however, the rate of change of concentration within a time unit t is assumed to be small compared to the source strength and to advection by the mean velocity, $\bar{u}$, an expression of the form

$$\chi = Q/A(H)\ \bar{u}t \tag{9a}$$

results where t is a unit of time (Fox, 1975). Variations of equation (9) have been used extensively in the literature on urban diffusion. Gifford has been most persistent in comparing such a technique to much more sophisticated models with successful results (Gifford, 1973). It has been shown (Fox, 1974), however, by using a rather limited but highly detailed data set from St. Louis, Missouri, that the accuracy of a computation increases as the sophistication increases. A comparison with experimentally measured concentrations yields standard deviations of 0.35 for the multiple-source gaussian diffusion model (Geomet, 1972), 0.39 for a vertically stratified box model, and 0.53 for a very sophisticated numerical solution of a finite difference version of equation (2) (Shir and Shieh, 1974).

The more direct numerical solution of various forms of equation (2) has been pursued in many papers. Basically, two techniques have been employed, one which essentially follows a mean wind trajectory by assuming a coherent parcel of air and allowing diffusion within that parcel as well as any appropriate chemical reactions (Eschenroeder, et al., 1972). This method has been applied extensively in Los Angeles, California where some recent field experiments have tentatively suggested that the assumptions of a coherent parcel of air may be quite

reasonable (Angell, et al., 1974). Liu and Seinfeld (1974) have presented an extensive discussion of the errors involved and a critical comparison of the accuracy of the trajectory approach as compared to a more conventional finite difference solution of (2).

A few models attempt such a finite difference solution of equation (2). The successes of such models are rather remarkable (Roth, et al., 1974 and Shir and Shieh, 1974). There is little doubt that this approach will in time replace all others. However, there are a number of difficulties. Certainly, one major limitation is the need for a large computing budget and for a great deal of specific information about the area under study, about the sources of pollution in the area, and, of course, about the meteorology.

The need for meteorological information is particularly acute since it is required in very fine detail in space and time. Obtaining such data is extremely expensive, and as a result there is an increasing interest in calculating wind fields using a model of the urban atmosphere on these very fine scales.

5. LONG-RANGE TRANSPORT

Distances greater than the multiple source or urban scale present somewhat different and more difficult problems to the atmospheric scientist. It is more difficult on this scale to separate the diffusion process from the meteorology. It is more difficult to study, measure, or forecast the meteorological conditions on this range of scales. It is more difficult to identify and to model the chemical reactions occurring with less concentrated species over the longer times and distances involved. These problems exist in part because there has been relatively little work done on long-range transport and mesoscale meteorology.

A number of recent problems, however, have pointed attention to the need for knowledge of the interactions between long-range transport and chemical reactions. Among these is the observation of very high oxidant values in rural or semirural surroundings. The generation of fine particulate matter (sulfates) over long ranges, the observation of pollution from one country in another and of course the problem of acidic precipitation.

Among the various subdivisions of meteorology there are at least three which must be drawn together to study long-range transport of pollution in general, and acid precipitation in particular. The micrometeorologist must be able to provide the knowledge of turbulence, dispersion, and removal mechanisms, the cloud physicist must input detailed cloud behavior to determine the efficiency of precipitation in removing pollution and the large-scale dynamicist must provide a knowledge of the transporting mechanism. This is made somewhat more complicated by the fact that the micrometeorologist is not used to dealing on such large-scales, while the dynamicist must come down in scale from his usual activity.

Perhaps the most common method of dealing with long-range transport is through the use of trajectories. Classically, meteorologists have used the trajectory to determine the current or past location of an air mass. The concept is rather simple in that one assumes knowledge exists about the flow field, namely its structure and its intensity, and that a hypothetical "air mass" remains coherent and is moved according to the velocity field. This approach has been used rather extensively by the OECD project (Nordø, 1975) and by the Air Resources Laboratory of NOAA (Miller, et al. 1975), among others. It is appropriate, however, to point out some of the difficulties in applying a trajectory analysis.

A major complication of atmospheric circulation is that the air system does not move uniformly like the sloshing in a glass of water, but rather exhibits often totally different behavior as a function of elevation. The vast majority of atmospheric flow is horizontal or two dimensional (in terms, for example, of total kinetic energy of the circulation) but the horizontal flow is different from one layer to the next, giving rise to both direction and magnitude changes in wind with height, so called shears. After the sun has gone below the horizon, the layers of air nearest the ground may become totally disassociated from the larger scale flow aloft. This nonuniformity presents a tremendous problem in analyzing where pollution goes over a 24-hour period.

One would expect the trajectory method to be a fairly good representation of pollution transport for flat terrain where the only questions are, how deep is the pollution layer, i.e., what is the inversion height, and what is the distribution of pollution through this height. One complication of long-range transport is the vertical component of motion. Organized vertical motion fields associated with topography or with the presence of meteorological "fronts", while somewhat predictable, are difficult to include in a trajectory model. The random vertical motion associated with thermal convection (cumulus cloud buildup) is generally not considered in long-range transport. Another problem is the lack of specialized meteorological data for even a simple trajectory analysis. Collection of the proper amount of meteorological information is prohibitively expensive. Thus, one must proceed with what information is available, generally only the larger scale standard weather maps. Such data must be supplemented on scales of concern with some sort of meteorological model.

Modeling mesoscale meteorology -- the scale from say 10 to 200 km -- is currently one of the most active fields of meteorological research. The success of modeling large-scale general circulation problems (Smagorinsky 1962) and small-scale turbulence (Fox and Lilly, 1972) has led to attempts to bridge the gap between these two fields. A major problem beside the lack of data is the nonuniqueness of mesoscale situations. To deal successfully with mesoscale problems, a large degree of parameterization must be incorporated into any model. This parameterization is necessary to account for both larger and smaller scale phenomena which affect the mesoscale. The ever changing general circulation pattern must be imposed as large-scale boundary conditions.

Methods must be developed to allow the introduction of these changing boundary conditions into a running calculation.

One of the more fundamental difficulties in mesoscale meteorological modeling is knowing what equation to solve. The equations of fluid motion, the Navier-Stokes equations, are similar to equation (2) but with a velocity component $\underset{\sim}{V}$ replacing the C_α and the addition of a pressure gradient force $(1/\rho)(\partial P/\partial x_i)$ and a buoyancy force $\Delta\rho g/\rho$ (in direction of gravity). The equations also require the addition of an internal conservation of mass (density) statement in order to be solvable. Solutions, however, are unstable when the Reynolds number (equal to velocity times a relevant length scale divided by the kinematic viscosity, ν, where ν is the molecular diffusivity of momentum divided by the density of the fluid) is as large as it generally is in the atmosphere. This instability leads to an equation for a turbulent fluid requiring the same type of averaging process as that for concentrations. The proper specification of $\overline{u_i' u_j'}$ in terms of mean quantities (closure) is essential to calculation of proper mesoscale fields. A number of models have been proposed (Donaldson, 1973) but a great deal of work remains to determine which performs best under realistic conditions. These turbulence terms are also responsible for the vertical spreading of pollution, and as such are tied up in the determination of long-range transport even more directly.

While it is not our purpose to review all the current mesoscale modeling, we will briefly mention a few of the techniques. The most detailed work is that of Deardorff (1973) (cf. Fox and Deardorff, 1972) who, through the use of three-dimensional numerical solutions of equations for small regions of the boundary layer, has sought to understand the nature of small-scale atmospheric behavior and to verify various theoretical developments. Other modelers who have attempted to retain as much of the nonlinearity of the problem as possible are Anthes (1974), Kreitzberg (1975), Pielke (1973), Pandolfo and Jacobs (1973) and Liu et al. (1974), to mention only a few. They attempt to solve the motion equations themselves. Another approach is to assume a multiple-layered atmosphere but simplify each layer. The efforts of Lavoie (1972), Tingle and Dieterle (1975) are examples of this procedure which solves an altered set of motion equations. A useful approach is to interpolate between measurements, but in a mathematically efficient manner. Objective analysis techniques, as these are called, can be forced to strictly reproduce the exact data, or can be weakly constrained to enforce selected conservation criteria (Sasaki, 1970). Dickerson (1975) and Sherman (1975) have used this approach for pollution applications.

The presence of mountainous terrain presents great problems to long-range transport analysis. Wind in mountains is affected by the local surface conditions as well as by larger scale motions. A model based on data interpolation introduced by Anderson (1971) has been adopted and refined for application to mountains by Fosberg, et al. (1975). Figures 3 and 4 illustrate some of the results.

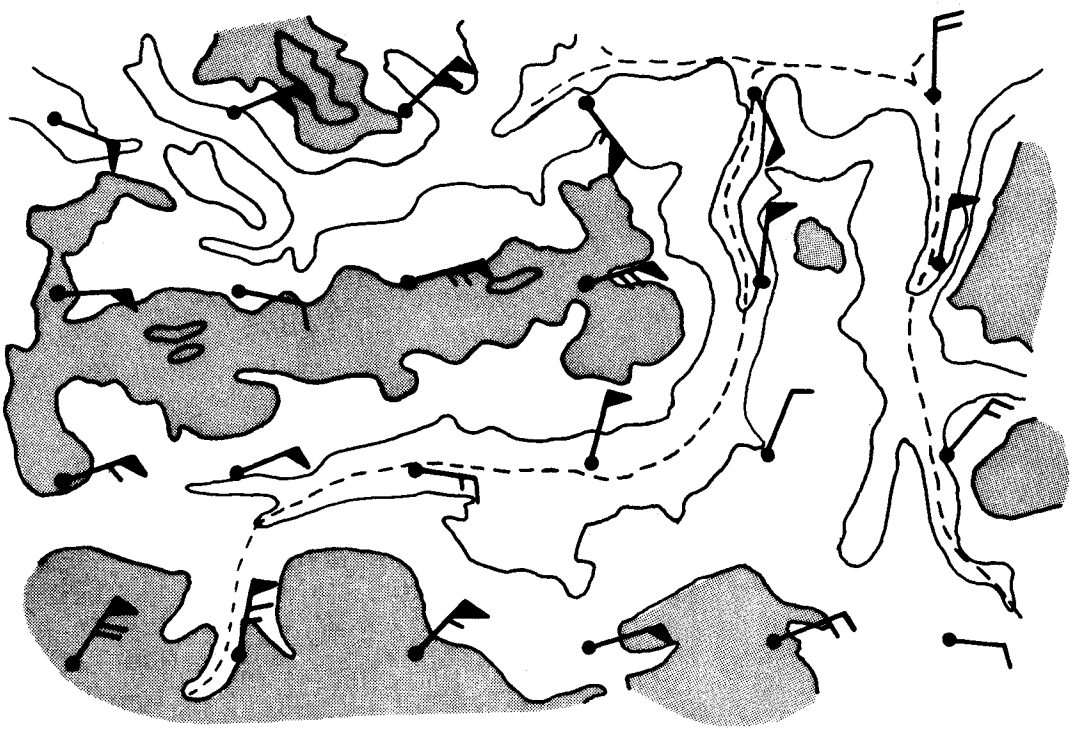

Figure 3. Windspeed and direction calculated for a mountainous area. Speeds are given by the sum of the barbs on the arrow tails. The arrow head points with the wind. Pennant is 5m/sec., full barb, 2m/sec., half barb, 1m/sec. (Fosberg, et al. 1975).

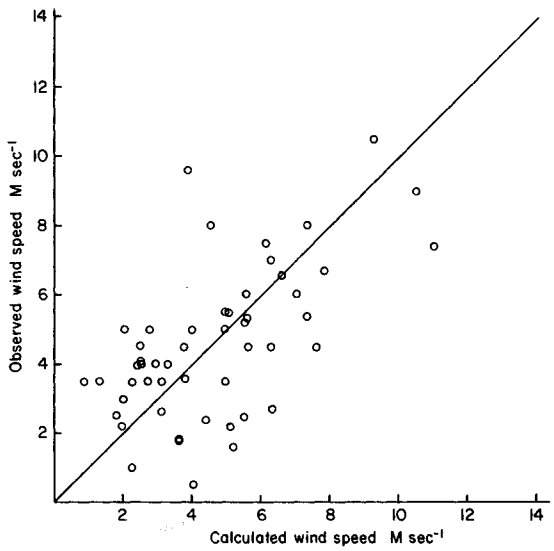

Figure 4a. Comparison of calculated windspeeds with observed windspeeds, (Fosberg, et al. 1975).

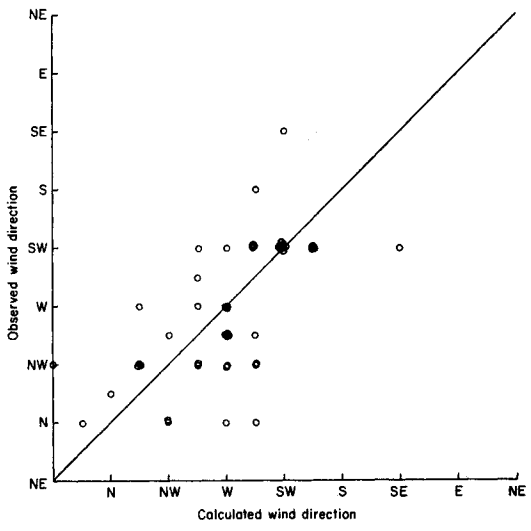

Figure 4b. Comparison of calculated wind directions with observed wind directions.

Finally, transport modeling was recently studied by Scriven and Fisher (1975). They present a simple moving box model which they show represents a good approximation of long-range transport. This technique is a basis for some calculations in the last section of this paper.

6. REMOVAL PROCESSES

It is difficult to deal with transport of air pollutants without considering removal. Removal occurs directly when the pollutant encounters the ground or any surface which has a sorbing capability for the pollutant, or indirectly when the pollutant becomes tied up in the precipitation cycle. Atmospheric chemistry plays a tremendous role in determining the nature of pollution removal alternatives.

Consider first the nonprecipitation or direct removal processes. Bolin, et al. (1974) and Chamberlain (1975) point out that deposition on a surface can be treated by considering a summation of resistances.

Resistance is defined as the inverse of the transport velocity (often called the deposition velocity) and is formally given by the expression

$$r = \frac{1}{V(Z_1, Z_2)} = \frac{\chi(Z_1) - \chi(Z_2)}{(F/\rho)} \tag{10}$$

where Z_1 and Z_2 represent the levels between which V and r are defined, χ is the concentration at each level, and F is the vertical flux (transport) of the concentration through $Z_1 - Z_2$.

Chamberlain (1975) suggests three separate resistances

$$r = r_a + r_b + r_c \tag{11}$$

where

r_a is a bulk aerodynamic resistance due to the structure of the air flow above the canopy,

r_b is due to the differences between mass and momentum transport,

r_c is the resistance due to the canopy.

r_a is determined from the velocity profile assumed as in Section 3 of the form $U = (U_*/k)\ln[(Z - d)/Z_o]$ where d is the displacement height required for the law to be used in the vicinity of large roughness elements such as trees. Analogous to equations (10) Bolin, et al.

(1974) considered the resistance to momentum transport as

$$V_m(Z_1,Z_2) = \tau/\rho\,[U(Z_1) - U(Z_2)] = U_*/[U(Z_1) - U(Z_2)] \quad (12)$$

where τ is the momentum flux or the stress, given by definition as ρU_*^2. The application of the logarithmic profile assumes $U(Z_2) = U(d) = 0$ so that

$$r_a = \frac{1}{V_m(Z_1,Z_2)} = \frac{U(Z_1)}{U_*} = \frac{1}{kU_*} \ln\left[\frac{(Z_1 - d)}{Z_o}\right] \quad (13)$$

Typical values for r_a over a forest can be determined by assuming $Z_1 = 10m$, $d = 7m$, $Z_o = .5m$, and $U_* \simeq .1 - .3$ m/sec. which yields $r_a \simeq 20 - 60$ sec./m.

The second resistance is to the transport of mass across the viscous surface layer. This resistance is expected to be dependent upon the viscosity of the fluid, the molecular diffusivity of the pollutant, and some shape factors of the surface. Chamberlain suggests:

$$r_b = B/U_* \quad (14)$$

where

$B \simeq 2(Re)^{\cdot 45}\, S_\alpha^{\cdot 8}$, for bluff resistance elements $Re = U_* Z_o/d$, $S_\alpha = \nu/\kappa_\alpha$

$B \simeq 7$ for SO_2 over fibrous roughness elements

The third resistance is due to the absorbing capability of the surface itself. There is some indication that r_c is related to stomatal resistance (Chamberlain, 1975; Kozlowski, 1974). Retention efficiency also seems to increase in the presence of high humidity. Experimentally determined values of r are listed in Table 1, from Chamberlain (1975). As Chamberlain points out, the values for $r_b + r_c$ are similar to the bulk surface resistance of crops to transpiration (minimum of 30 - 50 sec./m).

The removal of aerosol particles is a somewhat more complex problem, since the relative effects of Brownian motion, sedimentation, and particulate inertia must be assessed as a function of particle size. The impaction efficiency of the surface becomes highly important. The removal of aerosol particles must be assessed with a knowledge of both the physical and chemical nature of the aerosol and the surface configuration of the impactors. Chamberlain (1975) presents curves of particle size vs. deposition velocity and impaction coefficients (ratio of number of particles impacted to number of particles which would have passed through had the impaction surface not been there) as a function of cylinder diameter.

A forest canopy, however, creates a special flow field of its own.

Table 1. Resistances of canopies to uptake of SO_2 (from Chamberlain, 1975).

Method	No. of Expts.	Crop	u (1 m) m/s (mean)	u_* m/s (mean)	r $m^{-1}s$	$r_b + r_c$ $m^{-1}s$	Notes
Mass balance in crop		alfalfa	-	-	100	-	Growth chamber
		Rye-grass	-	-	160	-	Growth chamber
		Mustard	-	-	100	-	Glass-house (soil tagged with ^{35}S)
Mass balance in air	7	Alfalfa	3	0.4	40	-	Wind Tunnel, limited fetch over crop
Fumigation with $^{35}SO_2$	6	Grass	4.0	0.41	120	90	Field
Fumigation with $^{35}SO_2$	3	Grass	2.8	0.25	130	70	Field
Gradient	9	Grass	2.45	0.22	200	130	Field
Gradient	14	Short grass	3.0	0.18	360	-	Field
Gradient	19	Grass	3.0	0.29	220	180	Field
Gradient	13	Bare chalky soil	3.3	0.25	115	34	Field

Bergen (1975) has proposed a theory of momentum transfer through the forest canopy which suggests that the flow structure has a characteristic length scale on the order of the typical branch structure. Whether this branch structure (10-20 cm) or the actual needle structure (.1-1 cm) is the more significant for capturing small particulates, say below 10μ such as sulfate particles has not been determined. It is clear, however, that the increased roughness of forests compared to nonforested terrain will lead to an increased U_*, resulting in general with an increase in deposition velocity. Profiles of sulfate particles (Georgii, 1970) suggest, however, that the deposition velocity of such small particulate matter is at least an order of magnitude smaller than that of SO_2. Thus, most of the dry deposition of S comes in the form of SO_2 while sulfate particles are removed largely by precipitation processes.

As a final statement regarding dry removal, it should be emphasized that such a mechanism is operative whenever pollutants are present in the atmosphere. Wherever the surface wind shear stress is increased, deposition generally increases. Thus, forests should be expected to receive relatively larger amounts of pollutants from the atmosphere for two reasons: they create higher shear stresses because of both the trees and the general presence of elevated topography, and they present substantial amount of surface areas of sorbing material. Observation of this phenomenon has been presented in a number of papers in this symposium, including Knabe and Günther (1975) and Nyborg and Hocking (1975).

Removal by precipitation must be recognized as an intermittent process. A precise theory of the mechanics of removal is only part of the answer to precipitation removal. The other part is the occurrence or nonoccurrence of the precipitation itself, and of course the nature of that precipitation. Although a complete discussion of precipitation scavenging techniques is far beyond the scope of this review, we will briefly raise some comments. Recent meetings on the subject have been held (USAEC, 1974), volumes have been published that deal specifically with it (USAEC, 1970), and much work is still on going.

Pollution is removed by precipitation in two basic ways. Removal is generally termed "rain out" when the pollutant becomes involved in the precipitation-formation processes and "wash out" when it is simply caught by a falling drop. Rain out requires the aerosol to either act as a cloud condensation nucleus (CCN) or to be agglomerated into a CCN. Within the cloud, the condensation nucleus grows by a process of condensation and coalessence. Pollutants could well be introduced during this collisional buildup process. The relative importance of each process depends upon both the individual cloud and the cloud type. Wash out is a somewhat more straightforward process involving the simple collision of falling precipitation with the pollutant particles.

Dana, et al. (1973) have developed a model capable of determining the direct removal of SO_2 from plumes. This model indicates that even low background levels of SO_2 can significantly decrease wash out from a concentration plume, and that pH of rain has a strong effect on SO_2 scavenging. Indeed, the authors found situations where SO_2 in rain was decreased after passing through a plume. An additional observation of these studies is that sulfate was about five times more susceptible to wash out than was SO_2.

Rodhe and Grandell (1972) have used rainfall occurrence statistics to determine the removal rates of pollutants. Their study was oriented toward a calculation of the mean atmospheric residence time of aerosols in the troposphere, which they proved to be 100-300 hr. in summer and 35-80 hr. in winter. Their study was based upon the precipitation climatology of Sweden, and has not been applied to different precipitation regimes. Bolin, et al. (1974) calculated residence times by using the Rodhe and Grandell numbers in precipitation-scavenging and dry-deposition formulations in a simplified mass balance equation. They found that the ground parameters U_* and Z_o were most significant in determining residue times of pollutants emitted from tall stacks. Scriven and Fisher (1974), using an adaptation of a method suggested by Calder (1973), found a somewhat shorter residence time for SO_2 for approximately 24 hours. Any of these times, however, are probably sufficient to generate a good deal of $SO_4^=$, which contributes to the acidity of rain.

7. THE U.S. PROBLEM IN PERSPECTIVE

A number of the papers at this conference are oriented toward local problems or, at best, regional problems. The European work, however, does include significant considerations of the longer ranges of transport. To develop a base of understanding, it seems appropriate to consider the problem in the United States as a whole. To attempt such a broad and nonspecific inquiry we have purposely chosen rather broad and nonspecific modeling techniques. One of the basic principles of modeling is that no one component of the model is any more detailed and precise than any other component. We have made a number of gross assumptions which we will explain and attempt to justify in the next few paragraphs.

First, we have limited our consideration to SO_2 alone, although nitrogen-based anthropogenic emissions are responsible for as much as 30-40% of the strong-acid anion found in low pH samples in the U.S. However, characterization of NO_x emission is more complex than SO_2. Of the approximately 24 million tons of NO_x emitted annually, 45% is from industrial and power-generation point sources, while 36% is from transportation sources (U.S. EPA, 1974). In comparison, of the 32 million tons of SO_2 emitted, 87% are emitted through stationary point sources with the remaining 13% from stationary area sources. The complication of both active photochemical reactions and mobile sources make an analysis of NO_x pollution levels very difficult.

The chemical reactions which change SO_2 to $SO_4^=$ are complex. Calvert (1975), in this symposium, has raised a number of points regarding the relative importance of homogeneous (gas phase) reactions and heterogeneous oxidation occurring on solid particulate catalysts and in aqueous solution. The gas phase reactions apparently may be significant in low-concentration background atmospheres which have some NO_x and oxidizable organics available. One might speculate that a straightforward correlation between SO_2, NH_3, and water vapor could lead to an acceptable prediction of pH. Indeed in Scandinavia where extremely low pH is routinely found to be due to sulfates in solution (about 90% of the excess hydrogen ion is due to sulfuric acid) this approach works reasonably well, (Nordø, personal communication). In the U.S., however, this correlation does not seem to be as useful an approach because of elevated NO_x concentrations and the implications of NO_x involvement in both generation of $SO_4^=$ and direct formation of HNO_3. Thus, we have decided to calculate SO_2 concentrations assuming no chemical depletion, but accounting for dry and wet removal in an approximate manner.

Figure 5 is a map of the contiguous United States with the emission of SO_2 according to U.S. Environmental Protection Agency data (U.S. EPA, 1974), in each State. The choice of the political boundary of a State rather than the more reasonable air quality control region boundaries was based upon a desire to put no more precision into the calculation than it is worth. In considering a State such as New York, for example,

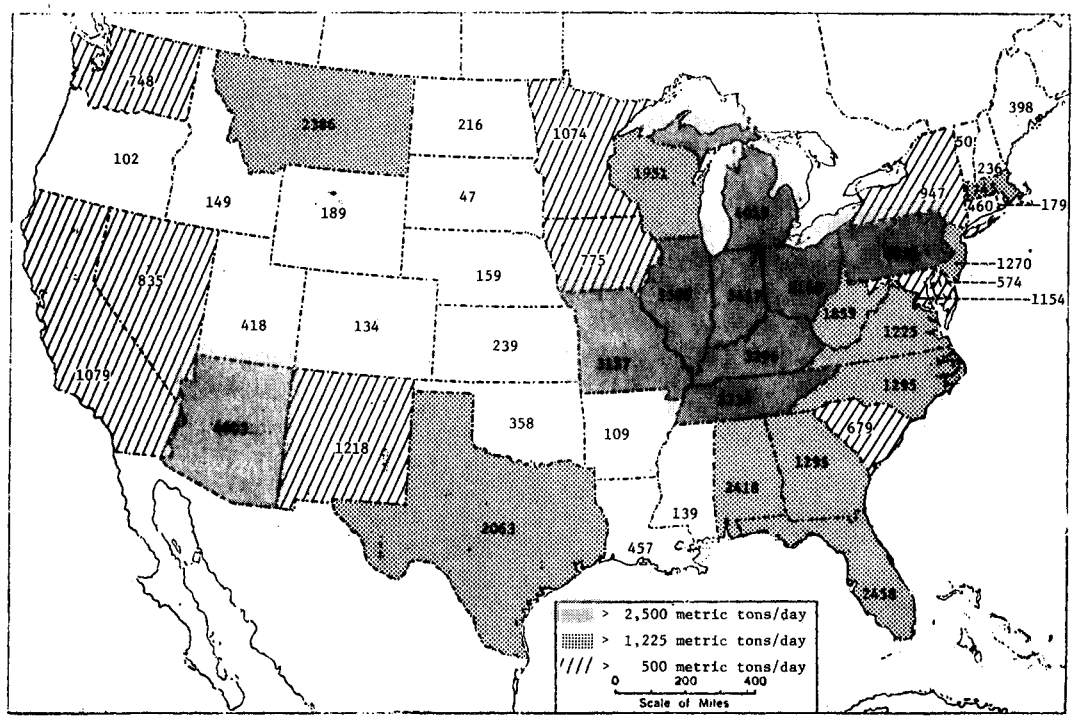

Figure 5. Emission of SO_2 in metric tons (10^3kg) per day from U.S. EPA, 1974.

it is obvious that the air quality of the New York metropolitan area is substantially different from the general background value for the State. If we had broken down the analysis to individual air quality control districts, the gross box modeling techniques we use would have to be improved to consider spatial variations, and the climatological data would need to be substantially improved.

To calculate the concentration, we will assume a uniform ventilated box model. The question is, what volume of air should be assumed to mix with the emission to determine a meaningful concentration? Assuming a mass balance, the flux of material entering a State is given by

$$\chi_i \, U_i \, H_i \, L_i$$

where χ_i is the entering concentration, U_i is the average velocity through the mixing layer, H_i is the height of the mixing layer, and L_i is the length scale over which these values are operating. Assuming the emission within the State is zero, and the flux out of the State on the downwind side is

$$\chi_o \, U_o \, H_o \, L_o$$

then the concentration leaving the State would be, assuming no internal removal mechanisms,

$$\chi_o = \chi_i \left(\frac{U_i H_i L_i}{U_o H_o L_o} \right) + Q/U_o H_o L_o$$

or if assume $\chi_i = 0$,

$$\chi = Q/U_o H_o L_o \qquad (15)$$

which we will use as an estimate of the ambient concentration. L_o is a length scale we assume is the square root of the surface area. The mean velocity is taken from Visher's climatology (Visher, 1954) as an interpolation over the State. The mixing depth from Holzworth (1972) is the interpolated summer afternoon mixing depth. Figure 6 shows the results of this analysis.

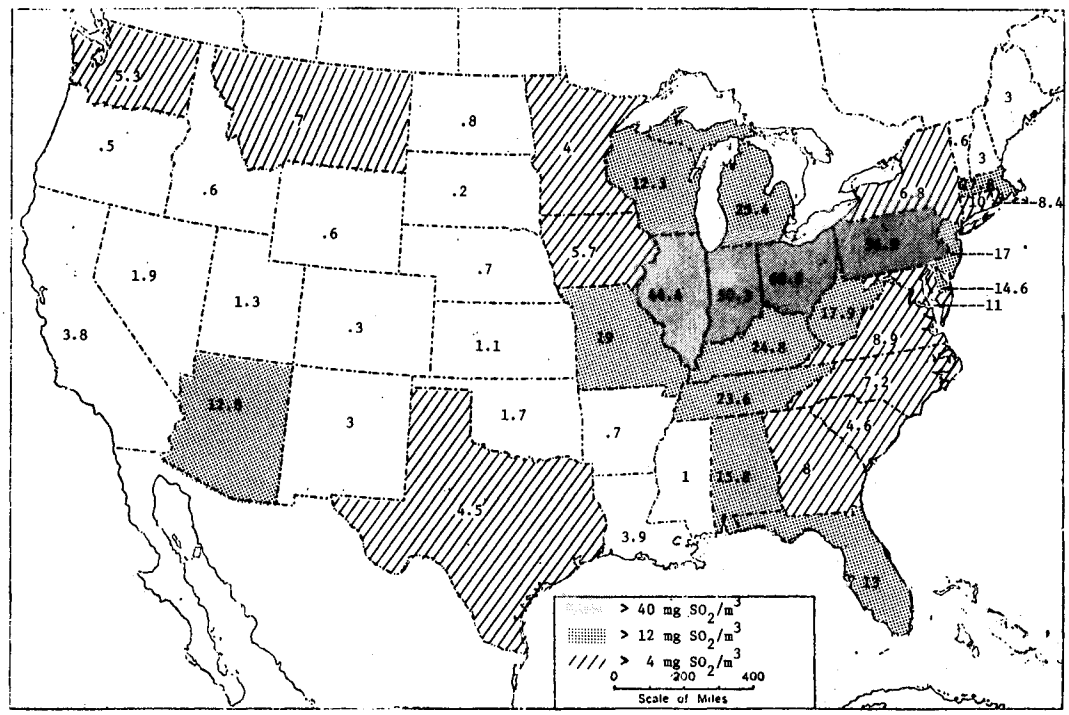

Figure 6. Concentration of SO_2 in $\mu g/m^3$ calculated using a simple ventilated box model, equation (15).

By assuming a mass balance between ambient concentration and dry deposition, Scriven and Fisher (1975) show that dry deposition, D is given by

$$D = \frac{V_g Q_o}{UHL_o} \exp\left(\frac{-Vgx}{UH}\right) = \frac{Q_o}{L_o x} \beta e^{-\beta}$$

which exhibits a maximum when $\beta = 1$ given by

$$D_{max} = Q_o / x_o L_o e$$

To estimate the maximum dry deposition over a State we have integrated this maximum over the entire State resulting in

$$D_{max} = [2\ Q_o / eL_o]\ \ln(L_o/2) \qquad (16)$$

Figure 7 shows the maximum dry deposition over each State, calculated from this equation.

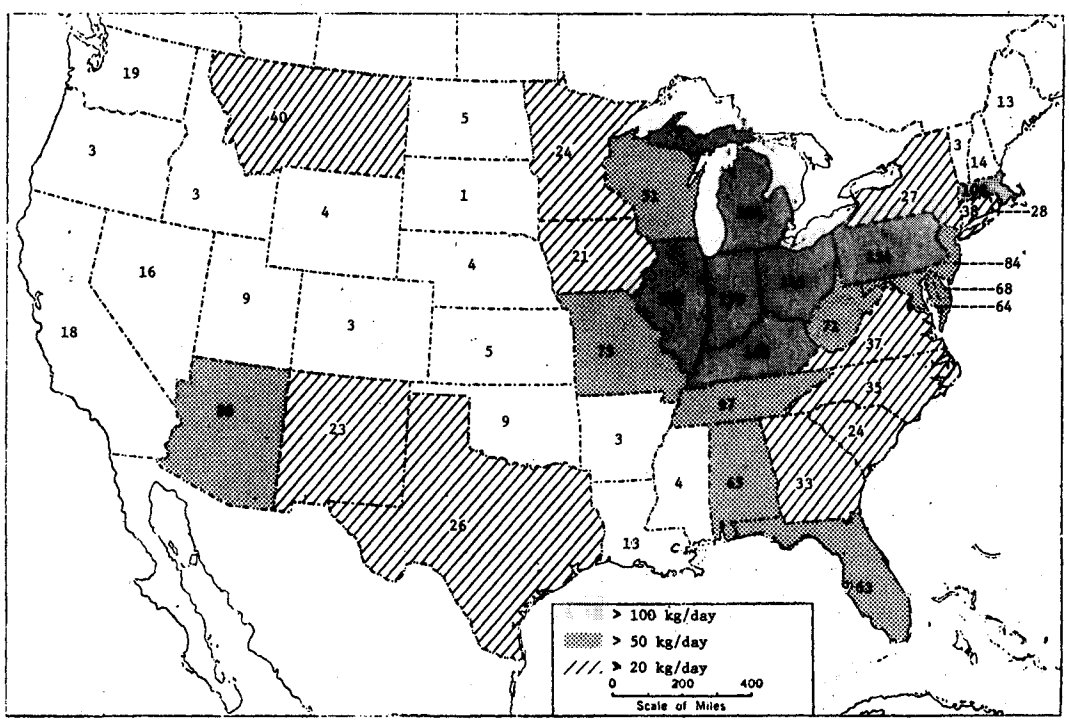

Figure 7. Maximum dry deposition in kilograms per day, calculated using equation (16).

To estimate the ratio of wet to dry deposition, we can again use the analysis of Scriven and Fisher (1975). They assume that removal by dry deposition is achieved by a deposition velocity Vg times the concentration χ, while precipitation removal is achieved by a "wash out factor" Λ related to the rainfall intensity. The removal term in a mass balance element under these assumptions becomes

$$(Vg + f\Lambda H)\ L_o\ \chi$$

One can then calculate the ratio of dry to wet removal r_{dw} by simply dividing the two components of this term, namely

$$r_{dw} = Vg/f\Lambda H \qquad (17)$$

where f is from Visher (1954) and represents the amount of time a rainfall of intensity i occurs, and Λ is the Langmur removal coefficient calculated according to Scriven and Fisher (1975) as

$$\Lambda \simeq 10^{-4}\ \sqrt{i}\ ;$$

where i has units of mm/hr. Figure 8 shows the results of the ratio of dry to wet removal, assuming that the deposition velocity is 1 cm/sec.

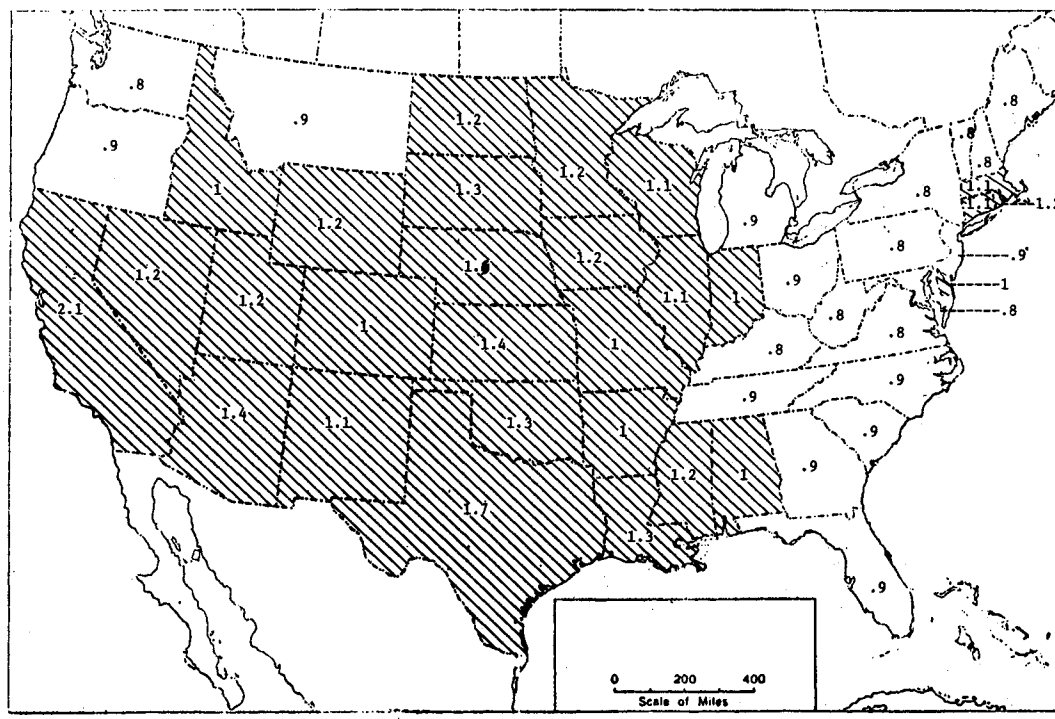

Figure 8. Ratio of dry to wet deposition, using equation (17) and assuming a deposition velocity of -1 cm/sec.

To improve the estimates of ambient concentration, we can use a mass balance model similar to Scriven and Fisher but including the contribution of sources within the transport distance. Considering a mass balance for a volume of width ∂x and length L_o, with the removal term as written above and the sources distributed as a general area source gives:

$$UL_o H \frac{d\chi}{dX} = -(Vg - f\Lambda H)\ L_o \chi\ Q_o/L_o$$

which is integrated to give

$$\chi(x) = \chi_o \exp[-(\frac{Vg}{H} + f\Lambda)\frac{x}{u}] + Q_o/HA(\frac{Vg}{H} + f\Lambda) \qquad (18)$$

where χ_o is the concentration at x = 0, the upwind edge of the area A. By assuming that $x = L_o/2$, an average distance to the center of the State, and that χ_o is an average of the concentrations in adjacent States, the ambient concentrations factoring in dry and wet removal and transport from other States can be calculated. Figure 9 shows the results of these computations. The significance of removal processes

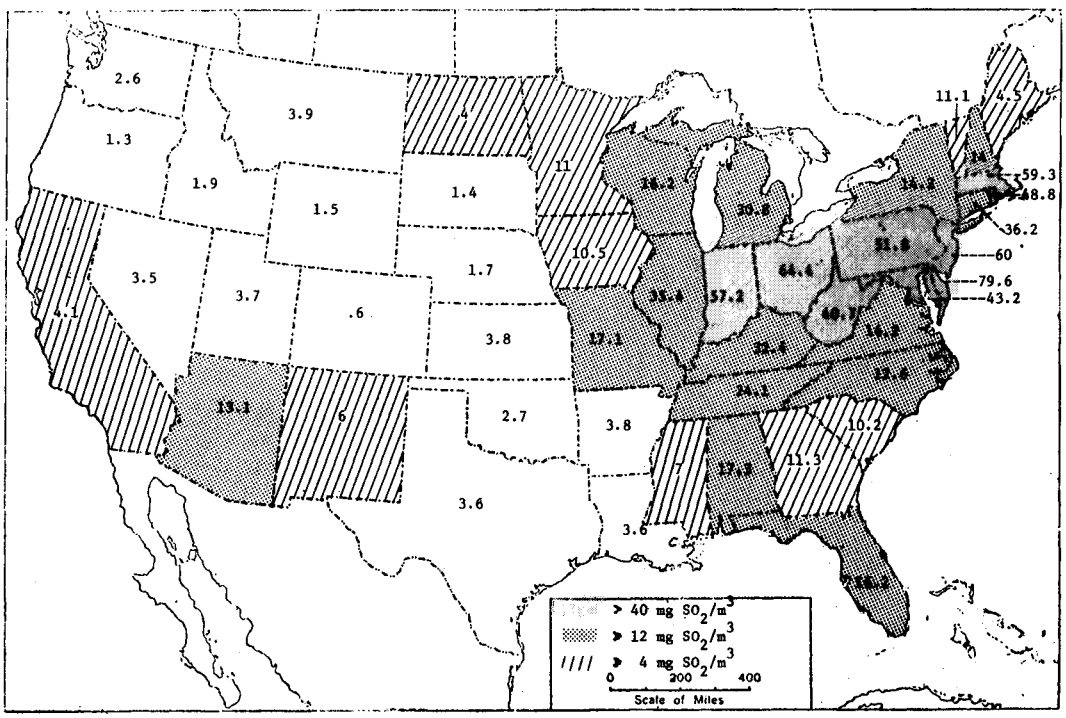

Figure 9. Concentration of SO_2 in $\mu g/m^3$ by considering effects of wet and dry deposition and transport from adjacent states, using equation (18).

and transport processes can be seen by comparing Figure 6 and Figure 9. Figure 6 estimates concentration assuming no removal and no out of state transport. Larger states removed from strong inflow of polluted air tend to show deposition processes are significant in reducing the concentrations. States in more heavily polluted areas are strongly affected by an influx of polluted air from adjacent areas.

It should be reiterated that these computations represent a gross estimate of the levels of SO_2 present in air over the United States. That they appear to present reasonable numbers is most likely due to the size of the area chosen for the analysis, and the fact that climatology is really not strongly variable from location to location. Furthermore, the computations point out that, except for some limited areas of the United States, particularly the Northeast, the vast majority of a State's ambient SO_2 comes from sources within the State.

Of concern, however, is the proposed coal development and utilization within the northern Great Plains. Recent estimates put the source strength at 2.5×10^6 tons SO_2/year which, emitted over an area of 5×10^{11} square meters, leads to a concentration increment of 14.8 $\mu g/m^3$. The impact of such an increase is certainly going to be significant to an area which currently has the cleanest air in the United States.

REFERENCES

Angell, J. K., Dickson, C. R. and Hoecker, W. H. 1974. RELATIVE DISPERSION WITHIN THE LOS ANGELES BASIN AS ESTIMATED FROM TETROON TRIADS, NOAA Technical Memorandum ERL-ARL-46.

Anderson, G. E. 1971. Mesoscale Influences on Wind Fields, J. APPL. METEOR., 10, 377.

Anthes, R. A. 1974. In Select Research Group in Air Pollution Meteorology Second Annual Report, EPA-650-14-74-045.

Bergen, J. D. 1975. In deVries, D. A. and Afgan, N. E., Ed. HEAT AND MASS TRANSFER IN THE BIOSPHERE, I. TRANSFER PROCESSES IN PLANT ENVIRONMENT, Wiley and Sons, New York, 287-298.

Bolin, B., Aspling, G., and Persson, C. 1974. Residence Time of Atmospheric Pollutants as Dependent on Source Characteristics, Atmospheric Diffusion Processes and Sink Mechanisms, TELLUS, XXVI, 185.

Briggs, G. A. 1969. Plume Rise, USAEC Critical Review Series.

Calder, K. L. 1972. Absorption of Ammonia from Atmospheric Plumes by Natural Water Surfaces, WATER, SOIL AIR POLLUT., 1, 375.

Calvert, J. G. 1975. Proc. First Int. Symp. on Acid Precipitation and the Forest Ecosystem, to be published.

Chamberlain, A. C. 1975. In deVires, D. A. and Afgan, N. H., ed., HEAT AND MASS TRANSFER IN THE BIOSPHERE, I. TRANSFER PROCESSES IN PLANT ENVIRONMENT, Wiley and Sons, New York, 561-582.

Dana, M. T., Hales, J. M. Slinn, W. G. N., and Wolf, M. A. 1973. Natural Precipitation Washout of Sulfur Compounds from Plumes, EPA-RS-73-047.

Deardorff, J. W. 1973. In Haugen, D. A., ed., Three-Dimensional Numerical Modeling of the Planetary Boundary Layer, WORKSHOP ON MICROMETEOROLOGY, Amer. Met. Soc., 271-309.

Dickerson, M. A. 1975. Abstract in Program First Conf. on Regional and Mesoscale Modeling, Bulletin of the Amer. Meteor. Soc., 56, 161.

Donaldson, C. du P. 1973. Haugen, D. A., Construction of a Dynamic Model of the Production of Atmospheric Turbulence and the Dispersal of Atmospheric Pollutants, Ed. WORKSHOP ON MICROMETERO-OLOGY, Amer. Met. Soc., 310.

Donaldson, C. Du P. and G. R. Hilst. 1972. In R. B. Landis and G. J. Hordemann, Ed. Chemical Reactions in Inhomogeneous Mixtures: The Effect of the Scale of Turbulent Mixing. PROC. OF THE 1972 HEAT TRANSFER AND FLUID MECHANICS INSTITUTE, Stanford Univ. Press, 253-261.

Eschenroeder, A. Q., J. Martinez, and R. Nordsleck. 1972. Evaluation of a Diffusion Model for Photochemical Survey Simulation, EPA-R4-73-012.

Fosberg, M. A., W. E. Marlatt, and L. Krupnak. 1975. ESTIMATION OF AIRFLOW PATTERNS OVER COMPLEX TERRAIN, to appear as Research Report, Rocky Mountain Forest and Range Exp. Station.

Fox, D. G. 1974. In Kornreich, L. D., Ed. The St. Louid Experience, PROC. OF SYM. ON CHEMICAL ASPECTS OF AIR QUALITY MODELING, Triangle University Consortium on Air Pollution, Chapel Hill, NC, 56-74.

Fox, D. G. 1975. Impact of concentrated recreational development, PROC. SYMP. ON MAN, LEISURE AND WILDLANDS, to be published.

Fox, D. G. and J. W. Deardorff. 1972. Computer Methods for Simulation of Multidimensional Nonlinear, Subsonic, Incompressible Flow, J. HEAT TRANSFER, 94, C. 337.

Fox, D. G. and D. K. Lilly. 1972. Numerical Simulation of Turbulent Flows, REV. OF GEOPHYSICS AND SPACE PHYSICS, 10, 51.

Geomet, Inc. 1972. Validation and Sensitivity Analysis of the Gaussian Plume Multiple Source Urban Diffusion Model, USEPA, OAP, APTD-U935.

Georgii, H. W. 1970. Contribution to the Atmospheric Sulfur Budget, J. GEOPH. RES, 75, 2365.

Gifford, F. A. 1974. Unpublished notes.

Gifford, F. A. 1973. The Simple ATDL Urban Air Pollution Model, PROC. OF THE FOURTH MEETING OF THE EXPERT PANEL ON AIR POLLUTION MODELING, NATO, CCMS.

Holzworth, G. C. 1972. MIXING HEIGHTS, WIND SPEEDS, AND POTENTIAL FOR URBAN AIR POLLUTION THROUGHOUT THE CONTIGUOUS UNITED STATES, EPA, OAP, AP-101.

Knabe, W. and Gunther, K. H. 1975. PROC. FIRST INT. SYMP. ON ACID PRECIPITATION AND THE FOREST ECOSYSTEM, to be published.

Kozlowski, T. T. 1974. In Reid, C. P. P., Ed., The Role of Educational Institutions in Forest Biology Research, PROC. OF THE THIRD NORTH AMERICAN FOREST BIOLOGY WORKSHOP, COLLEGE OF FORESTRY AND NATURAL RESOURCES, Colorado State University, 318-331.

Kreitzburg, C. 1975. Abstract in Program of the First Conf. on Regional and Mesoscale Modeling, BULLETIN OF THE AMER. METEOR. SOC., 56, 161.

Lavoie, R. L. 1973. A Mesoscale Numerical Model of Lake-Effect Storms; J. ATMOS. SCI., 29, 1025.

Liu, M. K., P. Mundkur, and M. A. Yocke. 1974. ASSESSMENT OF THE FEASIBILITY OF MODELING WIND FIELDS RELEVANT TO THE SPREAD OF BRUSH FIRES, Systems Application Inc.

Liu, M. K. and J. H. Seinfeld. 1974. In PROCEEDINGS SYMP. ON ATMOSPHERIC DIFFUSION AND AIR POLLUTION, American Meteorological Society.

Miller, J. M., J. Galloway, and G. E. Likens. 1975. In PROC. OF THE FIRST INT. SYMP. ON ACID PRECIPITATION OF THE FOREST ECOSYSTEM.

Nyborg, M. and D. Hocking. 1975. Ibid.

Nordø, J. 1975. Ibid.

Pandolfo, J. P. and C. A. Jacobs. 1973. Tests on an Urban Meteorological Pollutant Model, EPA-R4-73-025.

Pasquill, F. 1962. ATMOSPHERIC DIFFUSION, D. Van Nostrand, Inc. Princeton, NJ.

Pielke, R. A. 1973. A Three-Dimensional Numerical Model of the Sea Breezes over South Florida, NOAA Tech. Mem. ERL WMPO-2.

Rodhe, H. and J. Grandell. 1972. On the Removal Time of Aerosol Particles from the Atmosphere by Precipitation Scavenging, TELLUS, 24, 442.

Roth, P. M., P. J. W. Roberts, Mike Liu, S. D. Reynolds, and J. H. Seinfeld. 1974. Mathematical Modeling of Photochemical Air Pollution II A. Model and Inventory of Pollutant Emissions, ATMOS. ENVIRON., 8, 97.

Sasaki, Y. 1970. Some Basic Formalisms in Numerical Variational Analysis, MONTHLY WEATHER REV., 98, 875.

Scriven, R. A. and B. E. A. Fisher. The Long Range Transport of Airborne Material and its Removal by Deposition and Washout-I. General Considerations, ATMOS. ENVIRON., 9, 49.

Sherman, C. 1975. In Program First Conference on Regional Mesoscale Modeling, BULLETIN OF THE AMER. METEOR. SOC., 56, 161.

Shir, C. C. and L. J. Shieh. 1974. A Generalized Urban Air Pollution Model and its Application to the Study of the SO_2 Distributions in the St. Louis Metropolitan Area, J. OF APPL. METEOR., 13.

Slade, D. H., Ed. 1968. METEOROLOGY AND ATOMIC ENERGY, USAEC, Technical Information Division, Oak Ridge, TN, also as TID 24190, NTIS.

Smagorinsky, J. 1963. General Circulation Experiments with the Primitive Equations, p + 1, The Basic Experiment. MONTHLY WEATHER REV., 91, 99.

Smith, F. B. 1974. TURBULENCE IN THE ATMOSPHERE, unpublished report, British Met. Office, Bracknell, U.K.

Tingle, A. G. and D. A. Dieterle. 1975. Abstract in Program First Conference on Regional and Mesoscule Modeling, BULLETIN OF AMER. METEOR. SOC., 56, 161.

Turner, D. B. 1970. WORKBOOK OF ATMOSPHERIC DISPERSION ESTIMATES, EPA, OAP-AP-26.

USAEC. 1970. PRECIPITATION SCAVENGING, USAEC Tech. Info. Div., Oak Ridge, TN, also as CONF-700601, NTIS.

USAEC. 1974. PRECIPITATION SCAVENGING SYMPOSIUM, Champaign, IL, to appear from USAEC Tech. Info. Div., Oak Ridge, TN.

USEPA. 1974. 1972 National Emissions Report, National Emissions Data Systems of the Aerometric and Emissions Reporting System, EPA-450/2-74-012.

Visher, S. S. 1954. CLIMATIC ATLAS OF THE UNITED STATES, Harvard Univ. Press.

Stern, A. C., Ed. 1970. PROC. OF SYMPOS ON MULTIPLE-SOURCE URBAN DIFFUSION MODELS, USEPA, APCO Pub. No. AP-86.