Geographically-based Atmospheric Models

D. M. WHELPDALE

1 INTRODUCTION

This paper is an overview of geographically based atmospheric modeling, emphasizing regional and continental scales. Among the many applications of models, those most relevant to exposure assessment are the provision of media concentrations and areas of influence, and establishment of source/receptor relationships. In this context, the following features of modeling are reviewed: time and space scales, requirements for chemical information, the status of process knowledge, and data needs. The various types of atmospheric models are identified, and the two major long-range transport modeling approaches—Eulerian and Lagrangian—are described along with their advantages and limitations. A number of illustrative model applications are given to reference the use of atmospheric models in assessing exposure to chemicals in the atmosphere.

This paper comprehensively examines methods to assess exposures and their relations to human health and ecological injury by environmental chemicals. It is an overview of the current status of geographically-based (vs compartment) models, with emphasis on the larger scales. Topics for discussion include physical and chemical processes of transport, transformation and deposition; types of models, with their advantages and limitations; and illustrative applications demonstrating model use to assess receptor exposure.

Models provide a means to facilitate understanding of the behavior of a system. Atmospheric models are usually carefully structured sets of mathematical statements, but they may also be conceptual or physical (fluid tunnel). Emphasis here is on the first type. The main component of most atmospheric geographically-based models is the transport module, which provides the means of moving mass from one location to another. Other main categories of processes which may be included are chemical emissions; chemical, physical, and occasionally biological transformations; and those by which substances are removed from the atmosphere and deposited at the earth's surface.

Depending on the reason for their development and on their sophistication,

Methods for Assessing Exposure of Human and Non-Human Biota. Edited by R.G. Tardiff and B. Goldstein
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atmospheric models can have several applications:

(1) To provide a numerical result/description of a sequence of processes: e.g., the 24-h mean SO_4 concentration in air at a point downwind of a coal-fired generating station;

(2) To enable the synthesis or integration of knowledge of a series of atmospheric processes and, thus, to establish the status of our understanding, e.g., of what is known of the atmospheric pathways of PCBs from their sources of emission to their deposition rates to lakes;

(3) To aid in the interpretation of observations, e.g., in the use of back-trajectories associated with an episode of high O_3 levels to help identify precursor source regions;

(4) To interpolate between measurement stations, e.g., model calculations can be used to determine pollutant concentrations and deposition values at locations between network monitoring stations;

(5) To extrapolate (in space) or predict (in time), e.g., in models to predict the location of a pollutant cloud some hours or a few days hence;

(6) To aid in network or experiment design, e.g., to gain knowledge of pollutant emission fields and wind regime from a model to determine optimal sampling sites and station density; and

(7) To provide a theoretical framework for process research, e.g., with the more complex models to provide a means of integrating results of process research and of identifying gaps and weaknesses.

In the present context, the most relevant uses of models would include calculating (diagnostic or predictive) chemical concentrations in the vicinity of target organisms and of depositions rates (wet and dry) to these organisms, and describing pathways through the atmosphere, and the establishment of linkages between sources and receptors. It is important to note that atmospheric models usually "take" a pollutant to the immediate neighborhood of a receptor (i.e., ambient concentration). For pollutant removal by precipitation, the actual deposition rate to the surface of a lake or ecosystem is known; for dry deposition, the flux to the receptor surface is obtained by a parameterisation of the uptake processes—some of which are atmospheric while others are receptor dependent.

2 FACTORS INFLUENCING ATMOSPHERIC MODELING

2.1 TIME AND SPACE SCALES

Natural phenomena are often described in terms of their characteristic time or space scales. For example, meteorological phenomena are classified according to their characteristic horizontal dimensions with the terms micro, meso, synoptic, and global (Figure 1). The length of time that a chemical remains in the atmosphere, a characteristic parameterised by its residence time or lifetime (Rodhe, 1978), depends upon how effectively it is removed

by transformation and deposition processes. Both the meteorological and chemical aspects are important to determine the characteristics, scale, or impacts of an air pollution problem.

As an illustration of this interdependence, the key pollutant species in acid deposition are oxides of sulphur and nitrogen, which have average atmospheric residence times of a few days to a week. When these pollutants are emitted into the atmosphere where wind speeds are typically 10 m/s, they are transported and deposited over distances of a few thousand kilometers. This distance is the same as the extent of the heavily industrialised areas of Europe and North America, and a regional-to-continental scale pollution problem results in these areas. From an ecological perspective, the problem is potentially of a similar dimension; however, the impact can be modified by such factors as the distribution of sensitive receptors within a region.

The scale of a particular pollution problem is crucial to the correct choice of a model to be used for estimating exposure levels. Figure 1 shows the correspondence between time and space scales in the atmosphere and the four terms usually applied to pollution problems (Whelpdale, 1983):

(1) Local pollution problems are typically of a distance scale up to

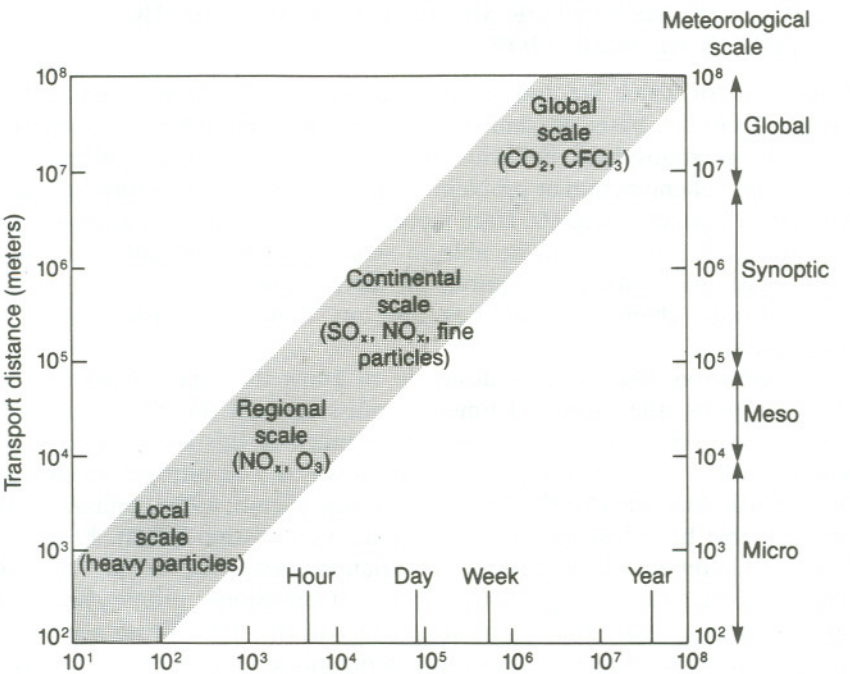

Figure 1. Characteristic air pollutions scales, shown with pollutant residence time and transport distance for a range of wind speeds

about 10 km and involve pollutants that are highly reactive (e.g., H_2S) or are rapidly deposited (large Pb-containing particles).

(2) Regional-scale problems may extend up to the order of 100 km and involve pollutants with lifetimes of several hours. An example is O_3, which is formed some distance downwind of sources of precursor emissions (NO, HC). Note that the definition of regional scale is somewhat controversial, depending, for example, on the size of one's country.

(3) Continental-scale problems range up to thousands of kilometers with chemical lifetimes of several days. The acid deposition issue has already been mentioned.

(4) Global-scale problems involve chemicals that are sufficiently long-lived to be transported throughout at least the hemisphere in which they are emitted, if not globally. CO_2 and perfluorocarbons are examples of this scale of problem.

In the following, models applicable to the regional and continental scales will be emphasised. Over the past three decades, much modeling has been done on smaller scales; whereas, for the global scales, geographically-based chemical modeling is in its infancy, and compartment models are still more useful.

2.2 REQUIREMENTS FOR MODELING ATMOSPHERIC CHEMICAL BEHAVIOR

Chemicals for which the atmosphere is an important exposure pathway are grouped on the basis of similarity of chemical and physical properties, similarity of atmospheric behavior, common emission sources, or being part of the same phenomenon or problem. Current examples are acidic pollutants, synthetic organics, nutrients, trace metals, radioactive species, and biological substances. In all cases, to be able to simulate their atmospheric behavior in models, information is required on their sources of emission, on their physical and chemical properties and on their transformation in the atmosphere.

For emission sources, it is desirable to know the types of sources and their locations, the rates and times of emission, and the characteristics of the sources. An example of a case where much of this information is well known is that of the primary acidic species, sulfur and nitrogen oxides. For both North America (MOI, 1982) and Europe (Semb, 1979), emission rates are available by industrial sector, location, and emission height. Progress is being made on establishing temporal variations (seasonal, weekly, diurnal), source characteristics (e.g., temperatures of emissions, primary/secondary species ratios), and estimates of uncertainty in the data. From a modeling perspective, these data are relatively well organised into emission inventories which are routinely used with acid deposition models.

For most other categories and substances, the situation is worse. For

individual chemicals, some of the information is known (e.g., the annual amount of DDT released to the environment may be approximated, but emission characteristics and locations are unknown); for some groups (e.g., metals), a systematic effort is underway to produce inventories (Cambell *et al.*, 1985; Pacyna, 1984); and for some (e.g., biological substances), almost nothing is known. Another example will serve to illustrate some of the difficulties (Seiber *et al.*, 1975). Pesticides may be released into the atmosphere during their manufacture, application, and disposal. In addition, they may revolatilise and be mobilised by wind-raised soil particles—perhaps several times. Whereas the former emissions are at least theoretically amenable to quantification, the latter are extremely difficult to manipulate. For most chemicals which circulate through the atmosphere, inadequate emission data are a serious constraint on reliable quantitative modeling of transport.

The physical and chemical properties of substances emitted into the atmosphere govern their initial disposition and subsequent behavior. Near roadways, particulate lead (Pb) has a bimodal size distribution. The larger particles fall out quickly, but those in the sub-micrometer mode are available for atmospheric transport over great distances. Whether a substance exists in the atmosphere in the gas phase, dissolved in cloud droplets, or adsorbed onto dust particles will affect its reactivity and removal efficiency, and thus the way its atmospheric behavior is simulated by a model. For several important organic chemicals in the atmosphere (e.g., pesticides and PCBs), their partitioning between gaseous and particulate phases is not well known.

Knowledge of chemical transformations is close second in importance to that of physical and chemical properties. Without understanding these processes, one is restricted to the use of either empirical or process-oriented models with highly simplified chemical parameterisations. Chemical modeling is well advanced in the fields of photochemical oxidants where successful applications have been made in urban areas such as Los Angeles. For acid deposition models, where the atmospheric chemistry of sulphur is relatively well known, linear parameterisations are still used. For most other chemicals, aside from the radioactive species, chemical transformations are handled very simply, if at all, in models.

To apply atmospheric models in a quantitative way to the assessment of exposure to chemicals, reliable information is required on emissions sources. To simulate atmospheric behavior of such chemicals, knowledge of their physical and chemical properties and of their transformations is necessary. For the majority of chemical species of interest, these data are unavailable.

2.3 ATMOSPHERIC PROCESSES

The atmospheric pathway between emission source and receptor consists of a number of processes which chemicals may experience: emissions, transport,

transformations, and removal. The realism with which these various processes are simulated in models depends upon both the depth of our knowledge of them and the scale of problem under consideration.

The authors do not intend to review the state of knowledge of all atmospheric processes, but rather to provide a few generalisations. For the scale of modeling being emphasised in this review (i.e., regional to continental), the processes associated with emissions are adequately known for most primary sources, even though locations and rates may not be. The physical processes of transport are reasonably well understood at the meteorological micro and synoptic scales; but meso scale processes involving orography, vertical exchanges and interactions with clouds continue to be difficult to model. As noted in the previous section, most physical and chemical transformations are not adequately understood to be realistically included in transport models. Local-scale photochemical oxidant models were noted as an exception earlier; and on a global scale, knowledge of transformation becomes less crucial. In the case of removal processes, they are sufficiently well understood to permit their simulation in a highly parameterised fashion in models; however, many details remain to be resolved.

As different models are applied to progressively larger scales, those processes important on a smaller scale must be adequately parameterised. On a local scale, processes associated with emissions, vertical motions, and highly reactive chemical species are important. On a regional scale, turbulent diffusion, exchanges between a mixed surface layer and the free troposphere, orographic lifting, lake-breeze, and other diurnal effects are also important. At continental scales, synoptic-scale atmospheric motions dominate, and wet deposition processes increase in influence. On a global scale, details of emissions and transformations have become less significant, while hemispheric circulation of longer-lived chemical species is of considerable importance.

From the viewpoint of human exposures to chemicals, two factors are noteworthy vis-à-vis scales: (1) wet deposition increases in importance relative to dry when going from small to larger scales, and (2) direct impacts of chemicals, usually related to ambient concentrations, give way to indirect effects, which are usually more closely tied to deposition, particularly for larger scales.

2.4 DATA REQUIREMENTS

The credibility of atmospheric models depends to a large extent on the availability of three types of data: (1) parameter values used in process simulations; (2) initial boundary conditions; and (3) evaluation data sets. Where models are used as conceptual frameworks to guide process research or as synthesizing tools, these requirements may not be stringent; however,

when used to guide control policy development or to serve in a legal context, these data—particularly evaluation data—become quite important.

The first category includes parameter values for chemical reaction rate constants, scavenging coefficients, and deposition velocities. For example, field measurements of the transformation rate of SO_2 to SO_4 yield values in the order of 10 percent/h in plumes and 0.1 percent/h in background air. In the current acid deposition models, values of approximately 0.7 percent/h have been found suitable (Eliassen and Saltbones, 1983) for annual averages. Newer formulations take account of seasonal variations in transformation rate: e.g.,

$$k_1 = 4\times10^{-6} + (2\times10^{-6})\sin(2t/T) \tag{1}$$

where T is one year (Eliassen, 1984a). The parameter used to describe dry deposition, the deposition velocity, has frequently been assumed to have an average value of 0.8 cm/s for SO_2 (Garland, 1978). Only recently have model simulations started to take account of atmospheric stability, surface roughness, and surface chemical and physical properties. Although it is difficult to generalise about the status of knowledge of parameter values, it is fair to say that the most difficult areas are those in which direct measurements are not available to verify parameters, as is the case for intermediate chemical species production rates (e.g., NO_x), gas/particle partitioning (e.g., PCBs), or dry deposition (all chemical species).

The second most important source of data is the input for a model and the initial boundary conditions. Input data fields needed to run most regional and continental scale models include anthropogenic emissions, wind velocity, and precipitation. Some require natural emissions, radiation, cloud cover, relative humidity, mixing height, and concentrations of reactive species. Depending on the sophistication of a model, surface fields, mixed layer averages, or three-dimensional, or time-dependent fields may be needed. Some models, such as the complex Eulerian models, require initialisation fields—that is, concentrations of all relevant chemical species throughout the model domain and/or at the boundaries of the model. Such requirements are extremely demanding and rarely available. Because of the long history of meteorological observations, meteorological data fields are usually more readily available than are chemical data fields.

Evaluation data are used with model predictions to help establish model reliability, credibility, and uncertainty estimates. As noted earlier, legal and control applications require demonstrated validity of models as established by evaluation with measurements. Model evaluation may include sensitivity testing, module-by-module testing with limited process-oriented field measurements, or complete model comparison against measured fields for some part of the entire domain. Sensitivity testing is carried out by varying one parameter at a time over the expected range of values, to determine

the magnitude of change associated with processes and data. Data requirements are meant to ensure that the appropriate ranges of values are considered. Testing individual modules (e.g., the transport part of a model) usually involves field experiments designed for a specific purpose, or the selection of weather situations where other processes (e.g., wet deposition) do not interfere. A recent example is the use of inert perfluorocarbon tracers in the CAPTEX experiment (Summers and Olson, 1985) to evaluate the transport and dispersion of an acid deposition model over distances up to 1000 km in eastern North America.

The evaluation of entire models against comprehensive data sets is still in its infancy because of the lack of such data sets. Limited evaluations have been carried out using both surface air concentration and wet deposition data for acidic species in North America (Voldner *et al.*, 1981) and Europe (Eliassen and Saltbones, 1983). In undertaking such model evaluations, it is important to consider that the accuracy and representativeness of measurements must be established in order to assess the quality of results from a model. While most measurements are made on surfaces, models usually provide results as either vertical resolutions or as layer-average. Thus, in the design and acquisition of evaluation data sets, above-surface values are very important.

In summary, data are available for parameterisation, input fields, and evaluation of the better understood processes and chemicals. Much less information is available for most trace metals and synthetic organics. There is also a need for comprehensive data sets for evaluation of the more complex atmospheric models.

3 TYPES OF MODELS

3.1 HOW MODELS ARE CHARACTERISED

The number of atmospheric models either ready for application or under development exceeds one hundred. The terms used to describe them seem to be almost as numerous. Nevertheless, most are characterised by one or more of the following.

(1) *Objective:* Models may be classified according to concentration, deposition, dosage (usually for radioactive species), or trajectory.

(2) *Spatial Scale:* The terms used to describe the spatial scales of air quality problems (and less frequently the meteorological scales) are also applied to local, regional, continental, and global models. In addition, scale-related terms such as urban, inter-regional, and long-range transport are frequently used.

(3) *Temporal Resolution:* An important distinction is made between models used over periods of a few hours to a few days, and those used on

a seasonal, annual, or longer basis. The former are termed "events" models and the latter "long-term" or, sometimes, "climatological" models.

(4) *Frame of Reference:* Another major distinction is whether a model is based on a frame of reference fixed to the earth (or a source) [an Eulerian model] or on a frame of reference fixed to a moving air parcel [a Lagrangian model].

(5) *Complexity:* The complexity of the model may be in its mathematical approach—analytical, statistical, probabilistic, physical (mass-conservation)—or it may be in its conceptual approach—empirical, mechanistic, process-oriented.

A model may legitimately have more than one of these characteristics. For example, a long-range transport model might be episodic, Lagrangian, and used for deposition calculations. In addition, some models are hybrids in that they combine features of two basic types, such as, an Eulerian-Lagrangian hybrid, or a statistical-empirical hybrid.

In the following sections, a few of the more commonly used models are described, with their advantages, limitations, and applications.

3.2 LOCAL-SCALE MODELS

The most frequently used modeling approach for practical applications on small scale is the Gaussian plume model. It is applicable to a single source and, based on a hypothesis about the distribution of material within a plume, it helps to determine the direction materials travel, how rapidly they are diluted in the atmosphere, and how they are removed. The models have been used extensively for exposure assessment. Detailed guidance on the use of Gaussian plume model formulations for a variety of source and receptor configurations is available in several references, notably Turner (1969) and Pasquill (1974).

The traditional assumptions on which Gaussian and other smaller-scale diffusion models have been developed—steady-state meteorological conditions, homogeneous turbulence, and homogeneous underlying surface—are too restrictive for many practical applications. Complicating factors such as topography, structures, forests, coastlines, water bodies, and time-dependent boundary-layer structure have made adaptions to the Gaussian approach necessary, and have stimulated the development of entirely new techniques. These factors are described in some detail in an excellent recent review by Barr and Clements (1984).

Finally, on local-scale modeling, the use of physical simulation models in fluid tunnels should not be neglected. Although most useful for non-reactive chemical simulations, they are able to deal effectively with problems of stratification and complex terrain.

3.3 LONG-RANGE TRANSPORT MODELS

These models emphasise regional and continental scales, using the long-range transport models. In the past ten years, interest in the acid deposition phenomenon has resulted in intense efforts to develop and improve models on this scale. The state of the art has been reviewed periodically in several excellent papers (e.g., Bass, 1980; Eliassen, 1980, 1984b; van den Hout and van Dop 1981; Bhumralkar and Ruff, 1984; Fisher, 1984).

3.3.1 Eulerian Models

The Eulerian approach divides the model domain into a two- or three-dimensional arrays of grid cells, fixed with respect to the earth's surface. Advection, diffusion, transformation, and removal are simulated in each grid cell by a set of mathematical expressions. Ideally, initial conditions are specified for each cell, and new output data (such as emissions) are injected into the appropriate cells.

The major advantages of the Eulerian models are the following:

(1) Capable, within the limits of knowledge, of treating complex processes such as three-dimensional advection/diffusion combined with non-linear interactions between various chemical species;

(2) Operate in a geographically fixed grid in which input data fields, winds, emissions, and precipitation are defined;

(3) Model output refers to the same grid and, therefore, information on the geographical distribution is automatically provided; and

(4) Their potential sophistication makes them powerful tools for integrating knowledge of atmospheric process over the entire model domain.

The limitations of the Eulerian approach are:

(1) Their complexity demands large amounts of computer time, computer storage, and input data;

(2) Because of the high cost of running these models, they are practical only for infrequent runs of limited duration; and

(3) Undesirable computational dispersion associated with numerical integration of the advection equation exist.

The sophistication and comprehensiveness that the Eulerian approach promises make it very attractive for many modeling applications, such as regional concentration and deposition patterns, source/receptor relationships, and scenario analysis. These lead directly to assessments of target exposure, to control strategy development, and policy formulation. However, this promise must be tempered with the current limitations imposed by incomplete knowledge, insufficient input data, high operating costs, and with the recognition that the most sophisticated models of this group are not yet operational. Within the next year or two, these models will be used successfully in the data-plentiful regions of the northeastern USA and southeastern Canada for problems of acid deposition and photochemical

oxidants. Finally, an important value of this "process-oriented" modeling approach is in its serving as a framework for planning and for synthesizing knowledge used in process research.

3.3.2 Lagrangian Models

The Lagrangian approach uses a moving frame of reference which is fixed with respect to moving air parcels transported through geographical regions of interest in accordance with observed or calculated wind fields. Diffusion, transformation, and removal calculations are performed for the moving parcels.

Lagrangian models may be either source- or receptor-oriented. In a source-oriented model, the positions of puffs emitted consecutively from each source are traced as a function of time. In a receptor-oriented model, the pollutant content of an air parcel is followed until the air parcel arrives at a selected receptor point.

The basis for Lagrangian models is the computation of air parcel trajectories. Trajectories, either forward or backward in time from a point of interest and without the inclusion of the chemical aspects of the model, are valuable and often-used interpretive tool in themselves (Miller, 1985). The main advantages to the Lagrangian approach to modeling are the following:

(1) Air parcels moving in the synoptic wind field are followed in a numerically simple manner, thus avoiding the computational dispersion of the Eulerian approach;

(2) They can be used to estimate contributions from individual sources or at individual receptors; and

(3) They are relatively easy and inexpensive to run on a computer.

The disadvantages of this approach include:

(1) Extension to three dimensions is difficult because of problems in simulating shear and diffusion;

(2) Most processes are highly parameterised; and, at least in the source-oriented model, non-linear chemical processes cannot be incorporated; and

(3) Errors are introduced when model output is interpolated back onto a geographically fixed grid.

Lagrangian models have been used extensively over the past 10 to 15 years for problems of regional-to-continental scales. As such their practical advantages and limitations are reasonably well known. The most attractive features of these models are their application to determine source/receptor contributions and their computational efficiency. They have been used extensively in Europe (Eliassen and Saltbones, 1983) and North America (MOI, 1982) for constructing regional concentration and deposition patterns and source/receptor relationships for seasonal and longer time periods.

Lagrangian models have also been used for the development of emission

control strategies through the running of scenarios and optimisation routines (Young and Shaw, 1985; Shaw, 1985) in Canada. Also in the policy area, and as a direct result of the flexibility of the Lagrangian approach, a simplified version of the model used in the European Monitoring and Evaluation Program (Eliassen and Saltbones, 1983) has been incorporated into a simulation model of acid deposition effects at the International Institute for Applied Systems Analysis (IIASA, 1984).

3.3.3 Statistical Trajectory Models

Statistical trajectory models are a subset of the Lagrangian models. There are two types. The first makes use of large numbers of air trajectories, either forward or backward, whose results are statistically analyzed to determine average pollutant contributions. In the second type, climatological average values of parameters are employed to calculate concentration and deposition fields. In addition to the advantages listed previously for the Lagrangian models, these are particularly cost-effective for repeated computer runs using various input scenarios. They are, however, "average" models, not amenable to short-term application and lacking in physical detail. For example, in the acid deposition application, they are unable to cope with the episodic nature of the phenomenon.

3.3.4 Other Modeling Approaches

The modeling approaches described above, or combinations of them, are the most common and most useful for the majority of atmospheric transport and exposure assessment problems. Two additional approaches, which are more empirical in nature, deserve mention.

The first of these is straight-forward, but unfortunately often neglected. The examination of available meteorological data in conjunction with chemical measurements, using rather simple analysis tools—such as data stratification based on meteorological parameters (Munn, 1970), wind roses, pollution mass budgets —can often help to diagnose or interpret a problem. This technique is most useful on the local to regional scales, and has been used, to identify likely source regions of precursors of ozone which damaged white bean and tobacco crops in southwestern Ontario and of tobacco blue mold spores which caused epidemics in Connecticut in 1979 and 1980 (Aylor *et al.*, 1982).

The final technique, called receptor modeling, is relatively new (Stevens and Pace, 1984). It involves the comparison of measured particle morphology, elemental chemical ratios, and variability at a receptor site with the pre-determined "signature" of individual sources or source regions, by means of statistical techniques. It has been used (Rahn and Lowenthal, 1985) in an attempt to determine which source(s) regions contribute most heavily to

pollutant concentration and deposition at selected receptor sites in the northeastern United States. With further refinement and use in conjunction with more traditional modeling approaches, this may prove to be a very useful source-identification technique.

3.4 GLOBAL-SCALE MODELS

Beyond the time scale of a few days (e.g., 5) or distance of 4000 km, the types of models discussed above become less useful. This is due both to incomplete input data fields on these scales, and to the growth of errors in computation and process simulation. Two-dimensional (height, latitude) models have been used to study global distributions of S and N species (Rodhe, 1985). Although useful for estimating hemispheric budgets and checking the simulations of transport, transformation, and removal processes, they are not useful for assessing concentrations near the earth's surface or thus for assessing exposure. Full three-dimensional global models, the general circulation models (GCMs), have been developed to study meteorological aspects of global circulation, and although used for relatively long-lived species such as N_2O and CO_2, they are not yet useful for predicting the behavior of reactive or short-lived species. At the current stage of development of modeling, the assessment of exposure on the global scale is better accomplished with the so-called compartmental models.

4 EXAMPLES OF APPLICATIONS OF ATMOSPHERIC MODELS

The scientific literature abounds with examples of the use of atmospheric models to describe the transport and other atmospheric behavior of chemicals. Most of these, however, pertain either to local scale and criteria pollutants or to regional continental scales with acidic pollutants. In relatively few cases is exposure assessment the direct aim in their application. A number of examples are described below either to highlight current modeling approaches to exposure assessment or to identify those with potential for this application.

4.1 DENSE GAS DISPERSION

As noted earlier the applications of both Gaussian and non-Gaussian dispersion models to local-scale pollution situations are numerous (e.g., Turner, 1969; Barr and Clements, 1984), and need not be discussed further. One specific area in which interest has been growing steadily is that of accidental releases of dense gases. Models developed for this type of problem are used primarily to establish safe designs and operating procedures

for the transport and storage of flammable and toxic materials, such as LNG, ammonia, or chlorine. Another application, however, is the evaluation of exposure to a material whose adverse effects may not appear for several years after a release.

Special factors to be considered in modeling of dense gases include:

(1) They are often toxic, combustible, or have boiling points below ambient temperature;

(2) They disperse at ground level and, therefore, efficiently encounter surface receptors;

(3) Underlying topography (gradient flow) and surface characteristics (temperature—causing boiling) are critical; and

(4) Their mode of release (often pressurised) and subsequent behaviour (boil-off) are conducive to droplet formation, which is difficult to model.

Blackmore *et al.* (1982) have published an in-depth review of 15 mathematical models currently used to study dense gas dispersion. The review includes an evaluation of their formulation, their capabilities, and comparisons with experimental data.

4.2 REGIONAL-SCALE CLIMATOLOGICAL CONCENTRATION AND DEPOSITION

Machta (1979) has prepared a handbook of calculation procedures for climatological estimates of pollution variables—ground-level air concentration, deposition rate, and horizontal flux—at distances of about 100 to 1000 km. This approach is based on a simple statistical model which involves the dilution of pollutants into sectors radiating from a source. Wind direction and speed are treated probabilistically; vertical spread is described by a vertical eddy diffusion model; and losses are allowed to occur by transformations, radioactive decay, and dry and wet deposition. Worksheets are provided which set out input information and arithmetic steps to calculate the variables desired. Appendices explain the theoretical basis and limitations of these procedures.

This simple approach can be used to estimate average, seasonal, or annual concentrations and/or deposition rates, and likely maximum short-term concentrations or deposition rates and their relative probability of occurrence, to assess health risks or monitoring needs. It is a useful first approach for exposure assessment on this scale.

4.3 AIRBORNE POLLEN TRANSPORT

Lagrangian models are often used only in a meteorological mode (i.e., without chemistry) to produce forward or backward air parcel trajectories. Among the many applications of such trajectories are stratification of air and precipitation composition measurements, and potential source

identification (Miller, 1985). One particularly interesting application has been the identification of source regions of tobacco blue mold spores and other airborne pollens (Aylor *et al.*, 1982; Raynor *et al.*, 1983). In one case where airborne transport of blue mold spores to Connecticut, USA was suspected in 1979 and 1980, back trajectories were used to identify possible source regions. Knowledge of the life cycle of the spores along with supplementary dispersion calculations and forward trajectories enabled an iterative approach to establish likely source regions and times. This application demonstrates the usefulness of such models for estimating the origins of various airborne materials such as plant disease spores and aeroallergens.

4.4 DEPOSITION OF AIRBORNE POLLUTANTS TO THE BALTIC

Few modeling studies have yet evaluated the atmospheric transport of, and exposure to, trace metals and organic chemicals. However, Rodhe *et al.* (1980) have shown how even a simple atmospheric modeling approach, based on air and precipitation concentrations, along with deposition parameters, can provide a very useful assessment of the quantities of a variety of chemicals entering an ecosystem, in this case the Baltic. The authors calculated annual fluxes and an uncertainty range for the nutrients N, S, and P; the metals Ca, Cr, Cu, Hg, Ni, Pd, V, and Zn; the radionuclides ^{90}Sr and ^{137}Cs; and the organic compounds DDT and PCB. Such estimates provide a basis for comparison with other inputs to the system, and with amounts in the reservoir. This study showed that the atmospheric pathway for N, several of the heavy metals, and the radionuclides was potentially important. The approach could be used to advantage to evaluate in a preliminary way the input or exposure to a fairly homogeneous ecosystem.

4.5 ATMOSPHERIC DISPERSION OF RADIOISOTOPES

This example and the following one demonstrate the more sophisticated approaches to long-range transport modeling that are currently in use. The first shows how a Lagrangian puff trajectory model, called MESOS, can be used to simulate the transport and dispersal of radionuclides over distances of several hundred km (ApSimon *et al.*, 1985a).

The model has been developed for application to the European area bounded by 44° and 62°N, and by 10°W and 20°E, and uses extensive meteorological data bases for the years 1973 and 1976. As part of its development, MESOS has been applied to the Windscale release of 1957, and has shown satisfactory agreement with observations. It has also been used to study national radioactive emissions from several sites in EEC countries (ApSimon *et al.*, 1985b). The results have been used to generate

annual average levels of contamination in air, and on the ground by wet and dry deposition. As a further step, these results were used with population-distribution and food-production data to estimate collective doses arising from direct exposure and ingestion per unit released of each nuclide. To assess risks from accidents, cumulative probability distributions exceeding specified levels of contamination were determined at selected receptor points around each source for releases over periods of three hours to seven days.

4.6 ACID DEPOSITION MODELING

Modeling in the acid deposition field has been on a firm operational basis for several years in both North America (e.g., MOI, 1982) and Europe (Eliassen and Saltbones,
1983). From an exposure assessment point of view, the products of interest are air and precipitation concentrations, and wet and dry deposition. Although only sulfur is incorporated into operational models, work is progressing on applications with nitrogen (Bottenheim *et al.*, 1984) and trace metals (Pacyna *et al.*, 1984).

The acid deposition model used in the European Monitoring and Evaluation Program is a one-layer, receptor-oriented Lagrangian trajectory model (Eliassen and Saltbones, 1983). As input, it uses a European-wide inventory of sulfur emissions and routine meteorological observations. Comparisons between model and measurements for airborne SO_2 and SO_4 and precipitation-SO_4 show good agreement on an annual basis (r-values of 0.82, 0.85 and 0.70, respectively) but not on a daily basis—thus emphasizing the longer-term applicability of this type of model. These illustrations show the operational state-of-the-art of modeling on this scale that might be applied to exposure assessment.

4.7 SOURCE/RECEPTOR RELATIONSHIPS

A recent application of regional-scale models (which is oriented more toward control strategy or policy development than toward receptor exposure) is the establishment of linkages between sources of pollutants and specific receptor locations, so-called "source/receptor relationships." For example, it may be desirable to know the relative contributions by eight different source regions to wet deposition at an ecologically sensitive lake. In the absence of unique tracers from each source region, this cannot be established by measurement; models must be used. This approach has been applied during the Canada/USA discussions on transboundary pollution (MOI, 1982).

In this application, a matrix of coefficients is calculated, "transfer coefficients," which relate each source region to each receptor region. This may be done for concentrations and deposition, and on an absolute or

relative basis. Because this approach relies on an assumption of linearity in the atmospheric system, uncertainties are associated with it; however, to date there has been satisfaction with its use to determine relative contributions from a variety of sources.

4.8 SIMULATION MODELING

A final example of the use of models on these scales is the incorporation of an atmospheric transport model into a simulation framework which goes from energy- emissions policy to forest-soil pH (IIASA, 1984). This model is a policy-oriented tool in which emphasis has been placed on comprehensibility and relative ease of use. It is modular, interactive, and has a graphical output.

The idea behind this approach was to incorporate current knowledge of the following aspects of the acid rain problem into a tool which could be used to demonstrate possible effects of energy-related emissions on control policy. These are:

(1) Country emissions and pollution control options;

(2) Atmospheric transport, transformation, and deposition of the emitted pollutants; and

(3) The implications of deposition for forest soil pH, which is used as an indicator of impact.

This process is in its early stage of development. Although results from it are best used as guidance and to stimulate thought, it is potentially an extremely useful tool to use in assessing the long-term environmental consequences of policy decisions.

5 SUMMARY AND CONCLUSIONS

This paper has presented an overview of geographically based atmospheric models, with a view to identifying models and features useful for assessing exposure to various chemicals. A variety of model applications was presented, and several factors which bear on the development and intended application of the models were discussed. A number of model types were described to point out their advantages, limitations, and optimal uses. Finally, a series of example applications was given to demonstrate how models, particularly those on the regional or continental scales, might be applied to exposure assessment.

The main conclusions from this review are:

(1) Exposure-related applications are most advanced on the local scale (not discussed here), and least developed on the global scale; for intermediate scales some techniques are available, particularly for longer-term applications for acidic chemicals.

(2) For intermediate scales, Lagrangian trajectory models can be used to advantage for a number of exposure problems. The Eulerian models, although showing much promise for better representation of processes and space/time resolution, are not yet proven for routine applications.

(3) Relatively few models have been evaluated adequately to be used confidently for quantitative prediction; however, several are available for diagnostic use and to aid in interpretation of measurements.

(4) A major limitation of modeling chemical exposure on most scales is the paucity of information on chemical emission sources—locations, rates, source characteristics—and for reactive chemicals, on their atmospheric transformations.

(5) A second limitation to be borne in mind is the spatial and temporal resolution possible with the larger scale models. Because they rely on meteorological observations of typically > 100 km and 6 or 12 h spacing, improved resolution of exposure is possible.

(6) Empirical measurements must continue to play an important role even with the use of models for exposure assessment. Input and evaluation data are essential for effective model use; furthermore, the parallel or iterative use of models and measurements often enhances the utility of the models.

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