Regulatory Ozone Modeling: Status, Directions, and Research Needs

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The Clean Air Act Amendments (CAAAs) of 1990 have established selected comprehensive, three-dimensional, Photochemical Air Quality Simulation Models (PAQSMs) as the required regulatory tools for analyzing the urban and regional problem of high ambient ozone levels across the United States. These models are currently applied to study and establish strategies for meeting the National Ambient Air Quality Standard (NAAQS) for ozone in nonattainment areas; State Implementation Plans (SIPs) resulting from these efforts must be submitted to the U.S. Environmental Protection Agency (U.S. EPA) in November 1994. The following presentation provides an overview and discussion of the regulatory ozone modeling process and its implications. First, the PAQSM-based ozone attainment demonstration process is summarized in the framework of the 1994 SIPs. Then, following a brief overview of the representation of physical and chemical processes in PAQSMs, the essential attributes of standard modeling systems currently in regulatory use are presented in a nonmathematical, self-contained format, intended to provide a basic understanding of both model capabilities and limitations. The types of air quality, emission, and meteorological data needed for applying and evaluating PAQSMs are discussed, as well as the sources, availability, and limitations of existing databases. The issue of evaluating a model’s performance in order to accept it as a tool for policy making is discussed, and various methodologies for implementing this objective are summarized. Selected interim results from diagnostic analyses, which are performed as a component of the regulatory ozone modeling process for the Philadelphia–New Jersey region, are also presented to provide some specific examples related to the general issues discussed in this work. Finally, research needs related to the evaluation and refinement of regulatory ozone modeling, the characterization of uncertainty in photochemical modeling, and the improvement of the model-based ozone-attainment demonstration process are presented to identify future directions in this area. — Environ Health Perspect 103(Suppl 2):107-132 (1999)

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Background

For at least the last two decades, ozone and its related photochemical oxidants have been the most persistent and perhaps the most dangerous air pollutants in the United States. Despite expenditures of billions of dollars for implementing emissions controls, ozone air quality has not drastically improved and the National Ambient Air Quality Standard (NAAQS) for ozone, an hourly averaged concentration of 0.12 ppm, is routinely violated in extensive urbanized regions, as well as in suburban and rural areas across the country. Indeed, in 1990, over 133 million Americans were living in areas where this standard was violated. The multifaceted nature of the problem stems from the fact that ozone is a secondary pollutant: it is formed in the troposphere through nonlinear chemical reactions between precursor species (oxides of nitrogen and volatile organic compounds) that are emitted by a wide variety of both anthropogenic and natural (biogenic) emission sources. The situation is exacerbated by meteorological conditions that can significantly enhance the ambient concentrations of ozone in specific locales.

The Clean Air Act requires the responsible state agencies to prepare an ozone attainment State Implementation Plan (SIP) for areas identified administratively as Metropolitan Statistical Areas (MSAs), Consolidated Metropolitan Statistical Areas (CMSAs), and counties where ozone levels do not comply with the NAAQS. This plan should document ways and means to meet and maintain ozone NAAQS in those areas in a time framework determined by the severity of the problem: for example, areas classified as having a severe ozone problem are allowed 15 or 17 years from 1990 to attain the ozone NAAQS, depending on their design value (see below), whereas areas classified as moderate are allowed 6 years. After approval by state and local governments and the U.S. EPA, the SIP is legally binding under both federal and state law.

The regulatory implementation of the standard requires that the average number of days exceeding an hourly averaged maximum concentration of 0.12 ppm over a 3-year period be less than or equal to one. This form of the ozone NAAQS allows for the occurrence of unusual meteorological events, which could result in more than one exceedance in any one year during a given 3-year period. So, if ozone monitoring sites in a region have recorded maximum hourly averaged ozone concentrations exceeding the 0.12 ppm level in 3 days during the first year while no exceedances were observed during the next 2 years, the region is considered to be in compliance with the ozone NAAQS. Depending on its ozone design value (DV), the fourth highest daily maximum hourly averaged ozone value monitored in the region over a 3-year period, each area in noncompliance with the standard is classified as a marginal (DV=1 10.121–0.138 ppm), moderate (DV=10.138–0.160 ppm), serious (DV=10.160–0.180 ppm),...
severe (DV > 0.180–0.280 ppm), and extreme (DV > 0.280 ppm) nonattainment area.

Because of the complexity of photochemical air pollution systems and due to a variety of interacting chemical and meteorological processes, the only rational way for quantifying the tropospheric ozone problem and for establishing causal relations among emission levels, meteorological conditions, and ambient air quality is through the application of comprehensive three-dimensional Photochemical Air Quality Simulation Models (PAQSMs). These models should incorporate in sufficient detail the current scientific understanding of the chemical and physical mechanisms affecting ozone formation, accumulation and transport. The Clean Air Act Amendments (CAA) of 1990 indeed require ozone nonattainment areas designated extreme, severe, and serious to demonstrate attainment of the ozone NAAQS through photochemical grid-based modeling. Intestate moderate areas are subject to similar requirements. Furthermore, intrastate moderate areas must demonstrate attainment through modeling, but use of grid models is an optional alternative to less comprehensive approaches. The SIPs that will contain enforceable regulations for ozone precursor emissions, based upon the results of the photochemical modeling, must be submitted to the U.S. EPA by November 15, 1994, after undergoing public hearings in each state.

The U.S. EPA has adopted the Urban Airshed Model, version IV (UAM-IV), an urban-scale grid-based model that has been under continuing development and refinement for almost two decades as the recommended model for photochemical modeling applications involving entire urban areas; other grid-based models may also be used after appropriate evaluation on a case by case basis. UAM and similar urban-scale models are typically applied to simulate photochemical air pollution dynamics over areas ranging from 50 x 50 to 300 x 300 km² with a grid resolution of 2 x 2 to 8 x 8 km² horizontally and 50 to 500 m vertically. For areas where long range transport of ozone and precursors is significant and affects air quality in multiple urban areas, a combination of regional and urban-scale modeling has been adopted by U.S. EPA and state environmental agencies as the preferred approach. Such an area is, for example, the Northeast Ozone Transport Region (NOTR), i.e., the metropolitan corridor extending from Washington, D.C., to Maine. The photochemical model used currently by U.S. EPA to assess regional dynamics and impact of ozone is the Regional Oxidant Model, version 2.2 (ROM-2.2); it simulates an atmosphere structured in three layers in the vertical direction and is applied over areas of 1,000 x 1,000 km² or larger, with a horizontal resolution of approximately 18.5 x 18.5 km². ROM has been applied to regional domains in the Northeast, South, and Midwest. The gross information calculated by ROM is then transferred to UAM or an equivalent urban-scale model, which performs a locally refined simulation in the form of initial and boundary conditions; this approach constitutes a so called one-way nesting of models, since the finer scale results are not utilized by the regional model. As an example, Figure 1 shows the modeling domain for the Philadelphia–New Jersey UAM-IV regulatory application, which includes the Philadelphia–Trenton–Wilmington Consolidated Metropolitan Statistical Area as well as the entire state of New Jersey, and utilizes a grid horizontally resolved at 5 x 5 km² embedded in the Regional Oxidant Modeling for Northeast Transport (ROMNET) project ROM2.2 domain. The structure of the comprehensive multiscale system, including core models, preprocessors, and postprocessors, employed for regulatory ozone modeling of this domain, is depicted schematically in Figure 2; sample time-series comparisons of interim surface ozone calculations for July 7 and 8, 1988, using this modeling system with values observed at selected monitoring sites, are presented in Figure 3. Such calculations represent part of a diagnostic analysis that is performed with the 1988 interim U.S. EPA emission inventory and various simplifying assumptions for the meteorological inputs (1).

Although PAQSMs have been under continuing development for over 20 years and have been applied extensively as both research and diagnostic tools, the 1994 ozone attainment SIP modeling process constitutes the first use of complex grid-based ozone models in a precise regulatory setting across the entire United States. This process, which has achieved the transformation of a research tool into a widely applied regulatory and policy tool, has also posed increased requirements for a) preparing more accurate emission and meteorological inputs for PAQSMs, b) valuating and interpreting model performance, and c) analyzing the consequences of modeling results. It has also accelerated the evolution of new models that are expected to improve the current standard, i.e., UAM-IV or the combined UAM-IV/ROM2.2 system. Currently, special programs, which are supported by extensive field studies and model development and evaluation projects, use an approach that is more integrated than that of the UAM-IV/ROM2.2 system. In these recently developed multi-scale models, the finer urban-scale grids are directly embedded within a coarser regional grid thus providing two-way grid nesting. When this approach is used, concentration estimates from the high resolution areas are used to calculate downwind concentrations in the low resolution areas. Such special programs, which are implemented within the regulatory setting of the 1994 ozone attainment SIPs, include the Lake Michigan Ozone Study (LMOS), and the joint San Joaquin Valley Air Quality Study and Atmospheric Utility Signatures, Predictions, and Experiments Study (SJVQAQS/AUSPEX) Regional Model Adaptation Program (SARMAP); the nested grid models utilized in these programs are, respectively, the Urban Airshed Model with Variable Grid (UAM-V) and the SARMAP Air Quality Model (SAQM), an adaptation of the Regional Acid Deposition Model (RADM).

The scientific issues underlying the physics and chemistry of the tropospheric ozone problem and their incorporation in air quality models, as well as many of the related policy questions, were summarized in 1991 in a report of the Committee on Tropospheric Ozone Formation and Measurement of the National Research Council (2); this document also provides an excellent introduction to the vast literature on these subjects. The following presentation contains a summary of the regulatory
modeling process, as defined in the framework of the 1990 CAAA and the 1994 ozone attainment SIPs. It also contains a nonmathematical overview and discussion of existing modeling tools, including photochemical air quality models and the necessary emissions and meteorological information preprocessor models, in the context of regulatory ozone modeling. The associated input and evaluation data needs, as well as the processes of applying and evaluating models in the ozone attainment demonstration framework, are also critically reviewed. Finally, issues related to research and database development needs for refining future ozone modeling practices are discussed.

The Regulatory Modeling Process

The regulatory application of grid-based PAQSMs, which, in most cases, involves either UAM-IV or the combined ROM2.2/ UAM-IV system, for the 1994 ozone attainment SIPs, affects a broad spectrum of society. The SIP modeling domains often encompass multiple geopolitical boundaries (counties, cities, and states) with a potentially large regulated community. Therefore, a regulatory photochemical application requires the coordination and collaboration of a large number of technical and policy groups to achieve a consistent modeling implementation, as well as a rational interpretation and use of the modeling results. Steps needed to conduct an urban-scale photochemical modeling study using the UAM or a similar PAQSM typically consist of the following (3,4):

- Establishment of a detailed technical protocol for the modeling study that will assign technical and policy responsibilities to all collaborating parties, identify technical and information resources, establish and revise time schedules, determine procedures for quality assuring databases, determine procedures for evaluating the validity of decisions made during the progress of the modeling project, etc.
- Identification of the appropriate boundaries of the domain to be modeled and of methods for determining initial and boundary conditions for air quality and meteorology information. The appropriate boundaries of the domain, in cases where the modeled area is not isolated, are typically determined as a compromise between the computational demands for enlargement of the domain and the level of error associated with boundary conditions considered acceptable for an application. Approaches for optimizing the selection of these boundaries include:
  - the examination of the degree of correlation among ozone monitoring stations as a function of distance;
  - the simulation of forward and backward airmass trajectories from selected locations (source and receptors) in the domain; and
  - the performance of simulations with the PAQSM over domains of decreasing size to directly determine the importance of domain size effects.
- Identification and preliminary comparative evaluation of alternative methods (preprocessor models) for managing and calculating detailed, episode-specific, emission and meteorological information.
- Selection of base-case historical ozone episodes (from 1987 to the present) to

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**Figure 2.** Structure of the comprehensive multiscale system, including core models, preprocessors, and postprocessors, employed for regulatory ozone modeling of the Philadelphia and New Jersey domain.

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**Figure 3.** Time-series comparison of interim surface ozone calculations for July 7 and 8, 1988, using the ROMZ.2/UAM-IV modeling system, with values observed at (A) an urban, (B) a suburban/industrial, and (C) a suburban/rural monitoring site. The high and low estimates from the four grid cells closest to the monitor are shown. The interim calculations represent part of a diagnostic analysis that is performed with the 1988 interim U.S. EPA emission inventory and various simplifying assumptions for the meteorological inputs.
be modeled for the attainment demonstration process; these episodes must be representative of the different meteorological regimes conducive to ozone formation in the area of concern.

- Acquisition and preprocessing (quality assurance, adjustment to day- and hour-specific temperature and activity, allocation to the appropriate grid cell, etc.) of air quality and meteorological and emissions data to develop the necessary input information for each meteorological episode that is to be used in the attainment demonstration model simulations.

- Application and component evaluation of major preprocessors (e.g., wind-field generators, emission models, etc.) for each selected meteorological ozone episode, using available field data.

- Application and evaluation of the complete modeling system, i.e., the photochemical model (UAM or other) with inputs prepared using (possibly more than one alternative) meteorological and emission preprocessors for each selected meteorological ozone episode.

- Implementation of diagnostic analyses on each meteorological episode simulation. The principal purpose of diagnostic analyses is to ensure that the model properly characterizes physical and chemical phenomena (wind fields and spatial and temporal emission patterns) instrumental in leading to observed ozone concentrations. The objective is to improve model performance, i.e., to achieve better spatial and temporal agreement with observed data. Diagnostic model simulations also uncover potential model input data gaps.

- Refinement and correction of inputs and input estimation methods, guided by the diagnostic analyses discussed above, and followed by the base-case application of the photochemical modeling system for each selected meteorological episode.

- Analysis of modeling results, using a series of graphical and numerical performance measures to determine overall model performance in replicating observed ozone concentrations and patterns. Model performance evaluation should also be done for ozone precursors (e.g., NO, NO\textsubscript{2}, VOCs) if suitable monitoring data are available. If the modeling system does not perform adequately for a past episode, even following adjustments and refinements suggested by the diagnostic analyses, then an alternative episode representing the same meteorological regime may be selected to replace it.

- For each meteorological episode selected, it is needed to estimate emissions as well as air quality for the projected attainment year required under the CAAA. Model simulations must be performed for each episode, implementing
  - projections of future changes in emissions and
  - mandated control measures (included in the CAAA), to determine whether the ozone NAAQS is expected to be met in the attainment year.

- If the model simulations for the attainment year do not show attainment for each modeled episode, it is needed to develop additional emission control strategies for selected source categories such as volatile organic compound (VOC) and/or NO\textsubscript{x} controls on selected source categories, alternative fuel scenarios, etc.

- Model simulations incorporating the aforementioned emission control measures are performed to demonstrate attainment of the ozone NAAQS for each meteorological episode. If the control measures do not show attainment, the previous step as well as this step, must be repeated as an iterative process until attainment is shown for each modeled episode.

In cases where there is significant inter-domain transport and regional and urban scale models are used in combination for the SIP modeling process (as is the case with UAM-IV and ROM2.2 for the modeling domains of the Northeast Ozone Transport Region), it is necessary to establish the efficacy of control strategies at a regional level. Thus, the regional model must also be applied in the final steps of the above process, incorporating the control measures identified in the urban-scale applications in order to study potential regional impact and interactions. Clearly, in situations like this, it is necessary to establish close and continuous collaboration among the modeling projects of adjacent domains to assure timely resolution of potential inconsistencies in both modeling approaches and policy recommendations.

It should be noted that the above regulatory application relies heavily upon the selection of a few representative historical ozone episodes for modeling. In principle, modeling simulations should be performed for every ozone episode during the ozone season (April to October for much of the United States) for an entire 3-year period to assure compliance with the ozone NAAQS. However, due to limitations in data, resources, and time, such an option is not viable. Consequently, the approach taken for the 1994 SIPs is to simulate a limited number of high ozone days and to require that the predicted maximum ozone concentration be less than 0.12 ppm at each grid cell in the modeling domain for all days simulated for the final emissions control strategy. Use of such an approach can be made consistent with the statistical form of the NAAQS only in a qualitative way through the choice of episodes that correspond to high ozone observations but that may not correspond to the absolute highest observations or control requirements (5). The episode selection process recommended by the U.S. EPA (3) is based on identifying meteorological regimes by constructing a climatological windrose of high ozone days. This process consists of the following steps: a) Establish days with ozone levels greater than the NAAQS; b) Determine the 7 to 10 A.M. resultant wind vector for all days chosen in step a and allocate them into eight compass directions and calms; c) Establish the Predominant Wind Directions (PWD) based on the maximum counts in one of the eight compass directions or calms; d) Assign the episode days based upon their resultant wind vectors to the PWD or other category and rank-order them based upon the observed ozone concentrations.

In practice, the above procedure may be appropriate for isolated urban domains having a single representative meteorological station rather than for extended urban areas with complex topography (involving coastlines or valleys) or involving regional transport. For such areas, the PWDs may be significantly different from meteorological station to station, and lumping different stations together may not be appropriate. An alternative approach that has been adopted in SIP modeling domains of the NOTR is to consider synoptic scale wind patterns to identify regimes associated with high ozone occurrences. For example, in the New Jersey–Philadelphia domain (4), ozone episodes from 1987 to 1991 were classified in five synoptic meteorological regimes (6) corresponding to a) prevailing S/SW winds (the majority of episodes); b) high pressure system above the domain — no significant transport (very few episodes); c) high pressure N or W of the domain (few episodes); d) frontal boundary within the domain (some episodes), and e) other, more complicated, meteorological conditions (some episodes). Other examples
of meteorological typing schemes are described in Horie (7), Zeldin (8), and Kalkstein et al. (9).

After the meteorological regimes have been determined, episodes are ranked according to various criteria such as observed ozone maximum, minimum, and average values within the domain, duration of the episode, spatial extent (pervasiveness) of the episode according to the fraction of ozone monitors within the domain in exceedance of the standard. Then episode days are selected from among the three highest ranked episode days from each meteorological regime. According to U.S. EPA Guidance (3) the primary modeling day for the attainment demonstration for each meteorological regime may be chosen to include any of the three top ranked days in that regime. In addition to considering the magnitude of the highest observed daily maximum ozone concentration in making this choice, data availability and quality, model performance, availability of regional modeling analyses, pervasiveness, and frequency with which observed meteorological conditions coincide with exceedances, are also recommended for consideration (3).

It should be mentioned that determination of the exact boundaries of the modeling domain and selection of the primary110 modeling days are not truly independent processes. Generally, it must be confirmed that the domain's downwind boundaries are sufficiently far from the CMSA/MSA that is the principal focus of the modeling study. This is done to ensure that emissions from the CMSA/MSA occurring on the primary day for each selected episode remain within the domain until 8:00 p.m. on that day. The extent of the upwind boundaries will depend on the proximity of large upwind source areas and the adequacy of techniques used to characterize incoming precursor concentrations. Large upwind emission source areas should be included in the modeling domain to the extent practicable. Also, if large uncertainty is anticipated for domain boundary conditions, the upwind boundaries should be located at a distance sufficient to minimize boundary effects on the model predictions in the center of the domain. Sensitivity analyses can assist in determining the effects of boundary conditions on calculated ozone concentration values (3,10).

**Photochemical Air Quality Data and Models**

**Air Quality Databases**

The attainment status of an area with respect to ozone is determined on the basis of monitored air quality data; from the perspective of regulatory ozone modeling, detailed ambient data on ozone and precursor (NO, and Volatile Organic Compounds (VOC)) concentrations are needed, both at ground level and aloft, to establish boundary and initial conditions for performing simulations and to evaluate the results of these simulations. Ground level concentrations of ozone are routinely recorded across the United States at National Air Monitoring Stations (NAMS) and State and Local Air Monitoring Stations (SLAMS). Some of these stations also monitor levels of oxides of nitrogen. Most of the monitors are located within or near urban areas, focusing on characterizing ozone impact on potential receptors. Some monitors are located in rural areas, with most having commercial or industrial activities nearby. There are only a few sites located in rural and coastal areas, although information from such sites is needed to characterize boundary conditions and long range transport (2,11). The 1990 CAAA also promulgated regulations for the enhanced monitoring of ozone and its precursors. In response to these regulations, the establishment of a network of Photochemical Assessment Monitoring Stations (PAMS) is currently under way (12). The PAMS network is intended to supplement the existing NAMS and SLAMS networks and to monitor criteria pollutants (i.e., for which NAAQS are established) as well as noncriteria pollutants. So PAMS will monitor O₃, NO, NO₂, and speciated VOCs, including several carbonyl species. The measurement of surface-level meteorological parameters such as wind speed and direction and temperature is required at all PAMS; furthermore, upper air meteorological measurements are required in some areas.

In general, currently available speciated ambient VOC data, as well as aloft air quality information, are very limited. Regarding ambient VOC speciation, U.S. EPA and state and local control agencies conducted a 3-year nationwide field study from June through September 1984, 1985, and 1987 at 47 sites in 41 cities to study the hydrocarbon composition in metropolitan areas (13). Three-hour integrated samples of hydrocarbons were taken at 0600 to 0900 LST from Monday through Friday on days forecasted to have high O₃ concentrations. All of the monitoring sites were located within metropolitan areas. These samples were analyzed using current techniques, and the specification results are considered valid. This is not generally true for samples collected and analyzed prior to 1982 due to numerous problems now known to be associated with older sampling and laboratory analysis procedures (11).

Useful information on the speciation and three-dimensional structure of ozone episodes has also been collected through various intensive field studies, implemented at various spatial and temporal scales and resolutions, that include among others the Northeast Regional Oxidant Studies (NEROS I and II) and the Persistent Elevated Pollution Episodes (PEPE) study (14), the Northeast Corridor Regional Modeling Project (NECRMP) (15), the Eulerian Model Evaluation Field Study (EMEFS) (16), the Southern California Air Quality Study (SCAQs) of 1987 (17), the Lake Michigan Oxidant Study (LMOS), and the SARMAB program. It should be noted however that although intensive field studies are valuable in understanding the dynamics of ozone episodes and in refining and evaluating models in a historic setting, they do not provide information that can be quantitatively extrapolated to other situations due to the large variations in meteorological conditions and in emission levels and composition from region to region and from year to year.

**Representation of Tropospheric Photochemistry in PAQSMs**

Ozone is formed in the troposphere as a product of a complex set of photochemical reactions of its precursors, i.e., oxides of nitrogen and reactive hydrocarbons such as aldehydes, alcohols, nitrogen and sulfur-containing organics. The terms ROG (reactive organic gases) and NMOG (nonmethane organic gases) are also often used as synonyms for VOCs. Excellent comprehensive treatments of the subject of atmospheric chemistry are available (2,18–21); a brief recent summary of the most important aspects of current interest can be found in Finlayson-Pitts and Pitts (22). Here, the overall cycles of organic oxidation and O₃, NO equilibrium that determine tropospheric chemical processes and ambient O₃ accumulation are summarized schematically in Figure 4.

PAQSMs incorporate chemical reaction mechanisms capable of predicting ozone and other secondary species levels resulting from inputs of organics and oxides of nitrogen. Gas-phase chemical reaction mechanisms for the atmospheric VOC/NOx system have, as a result of many years of smog chamber and laboratory kinetic studies,
reached a fairly advanced state of development. Available chemical mechanisms can be classified as explicit or detailed and as reduced or lumped. Explicit mechanisms aim to account for the detailed actual chemistry of each species and intermediate. Typically they involve several hundred reaction steps and are too lengthy to be incorporated in regulatory three-dimensional atmospheric models. For this reason, reduced or lumped mechanisms, generally involving fewer than 100 reactions, have been developed as systematic approximations of the detailed chemistry that is described by the explicit mechanisms.

Various methodologies have been used in the development of reduced mechanisms. Commonly, the inorganic chemistry that involves the NO/\text{O}_3/\text{HO} system is retained in full explicit form because of the relatively small number of inorganic species and reactions. The chemistry of organics is simplified by lumping together a number of reactions and chemical species. Two major approaches for performing this lumping can be identified. In the lumped molecule approach, the organics are grouped into classes according to their chemical character, e.g., as alkanes, alkenes, aromatics and carboxyls. Then either a generalized (hypothetical) species or a surrogate (actual) species is used to represent the chemistry of each lumped class. In the lumped structure approach the organics are grouped according to structure and reactivity characteristics. For example, carbon atoms may be grouped based on their bonding as single-bonded, double-bonded, or carbonyl.

It should be noted that, since the current photochemical mechanisms have multiple classes of reactive organics, the routinely measured or estimated total or reactive VOC concentrations and emissions must be converted into alkane-like or alkenelike hydrocarbon concentrations. This splitting or speciation process, which has to take into account source- and region-specific information, is very complex and constitutes one of the major sources of uncertainty associated with emission inventory development; it is further discussed in the Emission Data and Modeling section.

Differences among chemical mechanisms can be traced to different approaches to organic lumping and to different interpretations of kinetic and mechanistic uncertainties. At this time, the principal uncertainties lie in the detailed photooxidation pathways of aromatic molecules, such as toluene and the xylenes, in the reactions of the larger alkanes, in the photolysis and reactions of carbonyls, and in the photochemistry of biogenic VOC emission.

As a result of U.S. EPA-funded programs, two up to date chemical mechanisms for the formation of ozone in urban areas are presently available in the public domain. One is the SAPRC (Statewide Air Pollution Research Center, University of California) mechanism, also known as the CALL (Carter, Atkinson, Lloyd and Lurmann) mechanism, and the other is the CBM-IV (Carbon Bond mechanism, version IV, which is a condensed version of the more detailed Carbon Bond-X mechanism). CBM-IV is included in the regulatory versions of UAM-IV and ROM2.2. Both mechanisms have been tested against smog chamber data from the University of California, Riverside, and the University of North Carolina facilities. The predictions of each of these chemical mechanisms agree with these environmental chamber data to within about 30% for ozone maxima and show varying levels of reasonable agreement for other measurements and, despite somewhat different approaches, both can be judged to be acceptable for regulatory applications at this stage.

**Representation of Physical Processes in PAQSMs**

**Ozone Meteorology**

Although the production of ozone is a photochemical process, the rate and level of ozone accumulation in an area and its regional transport and impact are controlled by meteorological, (physical) processes. It is therefore critical that PAQSMs incorporate adequate representations of these processes, reflecting as realistically as possible the thermal and mechanical structure of the atmospheric boundary layer and the processes of advective transport due to the mean wind field of turbulent transport, the randomly fluctuating components of the wind field, and the deposition on the earth's surface. In general, all grid-based photochemical models require a description of the three-dimensional time-dependent wind fields and thermal structure together with information on ultraviolet radiation intensity, cloud cover, and moisture content of the atmosphere. Most ozone models also require a two-dimensional description of the mixing height over the modeling domain as a function of time. Information on dispersion rates is typically calculated from atmospheric wind field and atmospheric stability properties.

Before proceeding to discuss the representation of physical processes in PAQSMs, it is useful to summarize some qualitative features of ozone episode meteorology. Indeed, the occurrence of ozone episodes is associated with certain general meteorological features of the synoptic scale (1,000–5,000 km), regional scale (100–500 km), and local scale (10–50 km). These meteorological features must be incorporated in the air quality and associated meteorological preprocessor models.
through an adequate mathematical description.

Synoptic scale ozone episode meteorology is characterized by the presence of anticyclones, large regions of high pressure that produce clear skies, light winds, and a subsidence inversion. Such conditions further reduce vertical mixing, which can increase surface ozone concentrations. At the regional scale several types of meteorological features provide mechanisms for the transport of ozone and precursors from urban areas to rural areas or from one area to another area. For example, the nocturnal jet, a layer of fast (10 to 20 m/sec) wind at 200 to 500 m above ground level that forms after sunset, can transport ozone and precursors over significant distances at night. During the next day, surface ozone concentrations can increase as a result of the entrainment of this aged ozone and precursor air mass into the mixed layer. Another example relevant to coastal regions is the offshore/onshore flow which can occur under conditions associated with an anticyclone. During this type of flow, O and its precursors can be transported offshore where additional O can be produced without scavenging by fresh emissions of NOx. Then air masses with higher O concentrations can be transported onshore and impact a different region. Other situations of regional meteorology affecting ozone episodes may be associated with particular features of the terrain of the area (mountains, valleys, etc.). At the local scale, features such as sea breezes and venting of the boundary layer affect the accumulation of ozone. Land-sea breezes develop due to the temperature differences between land and water. During the day, warm rising air over the land is replaced by an onshore flow of cool air from over the water. The rising air diverges aloft, flows seaward, and sinks. At night the circulation reverses because the land is usually cooler than the water. More complicated flows can occur when a large scale background flow modulates the land sea breeze. As a result, land-sea breezes regularly can transport O and precursors aloft and over the water, eventually returning the photochemically aged air mass over land, with consequences similar to the regional circulation situation discussed above.

Venting of the boundary layer can occur when surface-based thermals become unstable and rise. If these thermals are warm enough to penetrate the subsidence inversion, the polluted air in the boundary layer is vented. During venting, cleaner tropospheric air can be transported downward into the boundary layer resulting in lower O concentrations. Cloud transport processes can also be important in ozone meteorology; strong thermal vertical updrafts, primarily originating near the surface in the lowest portion of the mixed layer, feed growing fair weather cumulus clouds with vertical air currents that extend in one steady upward motion from the ground to well above the top of the mixed layer. These types of clouds are termed fair weather cumulus since atmospheric conditions are such that they do not grow to the extent that precipitation forms. The dynamic effects of this transport process and daytime cloud evolution can have significant effects on the chemical fate of pollutants. For example, fresh emissions from the surface layer can be injected into a warm thermal and rise, essentially unmixed, to the top of the mixing layer, where they enter the base of a growing cumulus cloud. Within the cloud, the chemical processes of ambient pollutant species are suddenly altered by the presence of liquid water and the attendant attenuation of sunlight. The presence of fair weather cumulus clouds implies that the atmosphere above the earth’s boundary layer is too stably stratified for thermals to penetrate higher. In this case, the air comprising the tops of these clouds returns to the mixed layer and is heated on its descent, since it is being compressed by increasing atmospheric pressure. Ultimately, the air again arrives at the surface level where new emissions can be injected into it and ground deposition may occur, and the process may begin again. The time required for one complete cycle is typically 30 to 50 minutes with perhaps 10% of the time spent in the cloud stage.

The Structure of PAQSMs

Three-dimensional grid-based (also called airshed) PAQSMs describe pollutant transport, physical and chemical transformations, and physical removal in a fixed grid that is horizontally and vertically resolved in computational cells. Typically the horizontal dimension of the cell is in the range of 2 to 8 km for urban-scale applications and 10 to 100 km for regional-scale applications, while the vertical dimension typically ranges from 10 to a few hundred meters. In most regulatory ozone models only gas-phase chemistry is included, and this constitutes the only mechanism of pollutant transformation considered. Dry deposition is the only physical removal mechanism since ozone episodes are associated with fair weather conditions. In essence, the mathematical equations of a PAQSM represent a mass balance for each pollutant in each cell of the grid; they express the fact that a) the rate of accumulation of the mass of a pollutant species in each cell plus the net outflow rate of its mass, due to bulk or motion caused by the cell-average wind (advective transport) across the surface of the cell, must equal the sum of the net inflow rate of pollutant mass by turbulent dispersion across the cell surface; b) the net production or removal rate of the pollutant within the cell via chemical reactions, and c) the net removal rate of the pollutant via deposition. The equations describing this balance are discretized (are partial differential equations), as they express relations among rates of change and fluxes in three spatial dimensions and are coupled with each other through the chemical transformation terms; equations for adjacent cells are also coupled via the advective and turbulent transport terms. In practice, most numerical solution schemes assume some type of independence among operations in different spatial directions to decompose the partial differential equations into systems of ordinary differential equations. Even for a modest urban-scale application employing a 50 x 50 cell grid with six layers and the 35 species of the CB-IV mechanism, this requires a simultaneous numerical solution of a system of 525,000 (50 x 50 x 6 x 35) coupled differential equations over a time period corresponding to the episode under consideration. In cases of larger, multiscale domains or when multiphase chemistry and thermodynamic equilibria and physical transformations are considered, the numbers of simultaneous equations and the respective computational requirements rise dramatically. It should be noted that these computations do not include determination of the wind fields or meteorological parameters or the estimation and allocation of precursor emission rates for each cell. These tasks can actually be even more computationally demanding than the solution of the core model system. Traditionally, these tasks are assigned to so-called meteorological and emission pre-processors. These may constitute independent comprehensive modeling systems themselves that can be used in conjunction with various core photochemical models.

The accurate determination of emission rates for each species included in the model chemistry of an average wind vector for each cell of the computational grid for each time step of the numerical solution, as well as of the two-dimensional mixing height field over the modeling domain, are critical for a successful PAQSM application. These issues are discussed in the following sections under Emission Data and
Modeling, and Meteorological Data and Modeling.

Here we discuss briefly the treatment of turbulent transport and deposition that are typically included, along with gas-phase chemistry, in the core modules of photochemical modeling systems.

Treatment of Turbulent Transport in PAQSMs. Atmospheric turbulent transport (also referred to as dispersion, mixing, dilution and entrainment, depending on the application framework) is, for most practical purposes, uncoupled from the simultaneous transport of energy and momentum in the atmosphere, but still poses a difficult problem in modeling. Atmospheric applications require formulations that take into account stability variations, terrain complexities, and other attributes of the system modeled.

Research in turbulent dispersion has led to the development of many comprehensive theories that take into account the stochastic and nonlocal nature of the process. Both Eulerian (continuum or fixed-frame of reference) and Lagrangian (fluid particle) approaches have been applied and have given rise to models that range from simple similarity relations to parabolic and hyperbolic transport equations, to integrodifferential equations, to systems of stochastic ordinary differential equations, and to high order turbulence closure schemes involving systems of coupled partial differential equations (23). Nevertheless, despite the proliferation of alternatives in research models, the approaches commonly adopted by application-oriented models are rather limited. Indeed, in point source plume and puff models, turbulent dispersion is incorporated in dispersion parameters ($\sigma$) (24); in grid models it is described by a simple (local) gradient transport approximation that is parameterized with eddy diffusivities ($K'$s); the flux of mass is assumed proportional to the gradient of the mean concentration in a given direction, with $K$ being the proportionality factor. This is often referred to as K-theory of atmospheric dispersion. The resulting parabolic equation for pollutant transport is called the gradient transport or atmospheric diffusion equation (ADE) (19,23). Simple formulations for the $K'$s, as functions of atmospheric stability, are usually based on similarity theory. A variety of formulations are available and choice of a specific one usually relies on experience from previous use. Horizontal eddy diffusivities are usually assumed constant; formulas for vertical diffusivities incorporate a dependence on height. Typically, the vertical diffusivity increases (almost linearly) with height near the ground, remains almost constant at mid-mixing height, and decreases with height toward the top of the mixing layer.

It should be mentioned that atmospheric dispersion is affected by terrain complexity, presence of a coastline, cloud processes (entrainment and venting), and diurnal variations of boundary layer properties.

Treatment of Dry Deposition in PAQSMs. An important pathway for the removal of ozone and related pollutants from the atmosphere is dry deposition. Commonly, dry deposition models assume that the rate of removal of pollutants is proportional to their respective concentrations near the ground. The constant of proportionality (deposition velocity) depends on the pollutant as well as on meteorological and surface properties. In an attempt to incorporate more physical detail in this description, standard parameterizations utilize a linear resistance analogy for the removal process by modeling it as the result of three consecutive steps. Each step offers a resistance to the pollutant flux, and the deposition velocity is calculated as the inverse of the total resistance. These steps are (a) the aerodynamic step, in which pollutants are brought close to the surface by the action of turbulent dispersion; (b) the quasilinear sublayer step near the surface, where molecular diffusion becomes the dominant factor; and (c) the transfer uptake or surface step, which involves the actual removal of the pollutant by the surface through absorption, chemical reaction, or biochemical processes.

Calculating the aforementioned resistances is a complicated problem; many theoretical questions remain regarding the representation of near-surface mixing and the interactions of various pollutant species with the ground. However, some empirical parameterizations exist that provide values of the resistances in terms of land use information, surface roughness, atmospheric stability, season, and time of day (25).

PAQSMs for Regulatory Applications

UAM-IV (and its preceding versions) is the grid-based photochemical model that has undergone the most extensive usage and evaluation, with applications to several urban areas in the United States, Europe, and the Far East. It is also the most thoroughly documented model (3) and readily available in the public domain through U.S. EPA's Technology Transfer Network (TTN). UAM has been under continuing development since the early 1970s and currently represents the standard tool for urban-scale studies of photochemical pollution. UAM solves numerically the ADE in terrain-following coordinates on a three-dimensional grid covering the airshed of interest. This grid defines cells that are square in the horizontal direction and are typically of dimensions ranging from 2×2 to 8×8 km². In the vertical direction, the thickness of the layers of cells is determined by the diffusion break, the region top, and the minimum layer thickness. The diffusion break corresponds to the top of the spatially and temporally evolving mixing layer (either an unstable connective layer during the day or a shallow mechanically mixed layer at night). The region top is usually defined at or slightly above, the maximum daily diffusion break. Typical applications of UAM-IV employ 2 to 3 layers below and above the diffusion break, while the region top is located about 2 km above ground level.

The numerical solution methods used by UAM are based on finite difference schemes and employ the concept of fractional steps: (a) advection/diffusion is solved in the $x$-direction; (b) advection/diffusion is solved in the $y$-direction; (c) emissions are injected and advection/diffusion is solved in the vertical direction; and (d) chemical transformations are performed for reactive pollutants. The standard version of UAM-IV incorporates the CBM-IV photochemical mechanism. The maximum time step in the solution procedure is a function of grid size and the maximum wind velocity, which is determined to ensure numerical stability.

Although UAM has been refined significantly in the 20 or more years since its evolution, the fixed, horizontally uniform computational grid has retained its conceptual basis, until and including UAM-IV, the current regulatory version.

Development of ROM at the Atmospheric Sciences Research Laboratory of the U.S. EPA started in the mid 1970s, motivated by increasing recognition that summertime ozone episodes constitute regional phenomena. Since its first test release in 1983, ROM has undergone various changes and refinements; its current version, ROM2.2, is used by U.S. EPA to provide regional photochemical support for urban-scale, UAM-based modeling projects for the 1994 ozone attainment SIPs. ROM was designed to simulate most of the important chemical and physical processes responsible for the photochemical production of $O_3$ from emissions of natural and anthropogenic precursors over a domain of about 1000×1000 km² and for episodes of up to 15 days in duration. These processes include (a) horizontal
advective and turbulent transport; b) atmospheric chemistry; c) nighttime wind shear and transport associated with the low-level nocturnal jet; d) the effects of cumulus clouds on vertical mass transport and photochemical reaction rates; e) mesoscale vertical motions induced by terrain and large-scale flow; f) terrain effects on advection, diffusion, and deposition; and g) dry deposition. The processes are mathematically simulated in a three-dimensional grid-based model with three horizontal layers in the vertical direction. Horizontal grid resolution is 1/4° of longitude by 1/6° of latitude, or about 18.5 km × 18.5 km. The three model layers (bottom, middle, and top) are free to locally expand and contract in response to changes in the physical processes occurring within them. During the simulation period, horizontal advection, dispersion and gas-phase chemistry are modeled in all three layers. Concentration calculations in the bottom layer are used as estimates of surface concentrations. The time scale of output concentrations is 30 min, but ROM predictions are aggregated into 1-hr average concentrations. The bottom and middle layers of ROM represent the daytime mixed layer. Surface emissions are specified as a mass flux through the bottom layer. The modeling representation of the bottom layer incorporates a) the substantial wind shear that can exist in the lowest few hundred meters above ground in local areas where strong winds exist and the surface heat flux is weak; b) the thermal internal boundary layer that often exists over large lakes or near sea coasts; and c) deposition onto terrain features that protrude above the layer. At night, the middle layer represents what remains of the daytime mixed layer. As stable layers form near the ground and suppress turbulent vertical mixing, a nocturnal jet forms above the stable layer and can transport aged pollutant products and reactants at considerable distances. During the day, the top layer represents the synoptic subsidence inversion characteristic of high ozone concentration periods; its base is typically 1 to 2 km above the ground, and relatively clean tropospheric air is assumed to exist above its top at all times. If cumulus clouds are present, an upward flux of ozone and precursor species is injected into this layer by penetrative convection. At night, ozone and the remnants of other photochemical reaction products may remain in this layer and be transported long distances downwind. When cumulus clouds are present in a top layer cell, the upward mass flux from the surface is partially diverted from injection into the bottom layer to injection directly into the cumulus cloud or top layer. Within ROM, a submodel parameterizes the cloud flux process and its impact on mass fluxes among all the model's layers. The magnitude of the mass flux proceeding directly from the surface layer to the cloud layer is modeled as being proportional to the observed amount of cumulus cloud coverage and inversely proportional to the observed depth of the clouds.

For a ROM application, it is necessary to specify the initial and upward boundary ozone concentrations required to solve the governing equations of the model. Initial conditions are derived from estimates of mean tropospheric background concentrations. An effort is made to start ROM simulations at a time when O₃ concentrations are relatively low (usually several days prior to the period of particular interest when high concentrations occur). This strategy attempts to minimize the influence of uncertainties in the initial conditions on calculated concentrations during the period of greatest interest. The twice-daily (day-time and nighttime) gridded equilibrium concentrations for the 35 CBM-IV species used in ROM for the north, east, south, and west boundaries of each model layer are derived as follows: each boundary is assigned a single value for O₃ based on average ambient measurements at rural monitoring sites. The remaining 34 species are then equilibrated to this ozone concentration generating the set of concentration values for that boundary.

Land use input data consist of 11 land use categories for each ROM surface grid cell. These data are used to estimate biogenic emissions as a function of the area of vegetative land cover and to determine surface heat fluxes. Topographic data are also input and used in the calculation of layer heights.

A major limitation of ROM is that its resolution cannot be readily expanded in the vertical direction. The physical three-layer structure of ROM can present problems in mountainous areas where the atmospheric boundary layer has more than one level (2). Pollutants could be emitted in any or above the three model layers, depending on atmospheric stability and winds. In addition, transfer rates between layers can be affected by the presence of mountains whose heights are greater than the height of the boundary layer. Complications also may appear along coastal regions because of sea breeze circulations. Another concern is the ability of the model to adequately represent wind shear phenomena; it has been argued that this can affect conclusions concerning the relative effectiveness of VOC and NOₓ controls (11). Regional models providing higher resolution in the vertical direction should be able to alleviate many of these problems.

Other PAQMS for Regulatory Applications. UAM-IV is by far the most widely accepted PAQMS for urban-scale regulatory applications. Other urban-scale models have been used mostly as research tools. A potential alternative to UAM-IV is CALGRID, which has similar input data requirements as well as computational objectives, but uses different parameterizations and numerical solution methods; the chemistry incorporated in CALGRID is the SAPRC mechanism. There is however a variety of application-oriented regional scale models that incorporate tropospheric photochemistry. These include the Acid Deposition and Oxidant Model (ADOM) developed at Environmental Science Research (ESR); the Regional Acid Deposition Model (RADM) developed at the National Center for Atmospheric Research and at the State University of New York; the Regional Transport Model (RTM-III) developed at Systems Applications International; the Sulfur Transport Eulerian Model (STEM-II) developed at the University of Kentucky and the University of Iowa; the nested-grid Urban Airshed Model-V (UAM-V) developed at Systems Applications International; and the variable-grid Urban Regional Model (URM) developed at Carnegie Mellon University. ADOM, RADM, and STEM-II are regional oxidant and acid deposition models containing both gas- and aqueous-phase chemical mechanisms. RTM-III, UAM-V and URM, like ROM, are regional oxidant models focusing on gas-phase photochemistry.

It has been suggested (11,26) that regional oxidant models can predict observed ambient O₃ levels to within 30 to 40% on average. ADOM, RADM, and STEM-II have been evaluated using acid precipitation data sets. In particular, ADOM and RADM have been evaluated using the OSCAR database, and STEM-II was evaluated using the PRECP database. STEM-II has been evaluated in three more limited geographical areas, including the Philadelphia area, central Japan, and Kentucky. UAM-V has been applied in the South Coast Air Basin and the Lake Michigan area.

All models provide means for simulating pollutant transport using three-dimensional wind flow inputs. Furthermore, RADM,
ADOM, STEM-II, and ROM include provisions for treating the vertical redistribution of pollutants, resulting from the presence of cumulus clouds. UAM-V does not explicitly provide such capability unless the effects of cumulus clouds are embedded within the three dimensional flow fields generated by the meteorological preprocessor model.

Vertical turbulent dispersion is represented in all the above models using K-theory. The various parameterizations depend on atmospheric stability, and in some cases, other meteorological parameters. Horizontal turbulent dispersion is generally considered to be less important, especially for regional models with limited horizontal grid resolution. RADM and ADOM provide no representation of horizontal dispersion, and the other models include simple parameterizations based on wind speed, convective velocity scale, etc. The ability of the model to provide adequate horizontal grid resolution in areas where significant concentration gradients may occur (in the proximity of significant source areas) or to provide a subgrid scale representation of initial plume dispersion may be more important than the treatment of horizontal dispersion process (11). Of course, as the grid resolution increases, the horizontal dispersion process takes on greater importance, especially near source areas. UAM-V can simulate the emissions from large point-sources using a reactive plume-in-grid module, which maintains the integrity of the plume until such time as it attains a dimension commensurate with that of the grid cell. Such a module can potentially provide a more accurate estimation of the chemical transformations that occur in a point source plume and the effects of such emissions on air quality in areas downwind of the source.

It should be mentioned that modeling approximations reflecting the nonlocal character of atmospheric transport and mixing, such as the transient turbulence model or asymmetric convective modeling are currently gaining acceptance in the formulation of air quality models, such as the evolving versions of RADM.

All the above models incorporate gas-phase kinetic mechanisms treating chemical transformations involving NO$_x$, VOCs, and O$_3$. These are condensed mechanisms employing either lumped structure, Carbon Bond type (ROM, RTM-III, STEMI, or UAM-V) or lumped molecule (RADM, ADOM, or STEM-II) concepts. In general, the mechanisms reflect up to date experimental kinetic and mechanistic data.

RADM treats precipitating cumulus and stratus clouds and fair weather clouds. Cloud average values are used to treat cloud chemistry with a box model approach. ADOM treats cumulus clouds using a comprehensive representation of cloud dynamics but a simple treatment of cloud microphysics. The stratus cloud model includes a detailed treatment of cloud microphysics. STEM-II includes a module providing a detailed treatment of cloud microphysics. RTM-III and UAM-V provide no representation of cloud processes.

With regard to future developments, it should be mentioned that the Office of Research and Development (ORD) of the U.S. EPA has initiated a long-range plan for implementing a modular modeling system that will consolidate emission and meteorological input preparation methods and will provide state of the art data management, analysis and visualization methods. This third generation modeling framework, which is developed in conjunction with the Federal High Performance Computing and Communications initiative, is referred to as MODELS-3 (27). This modeling system will consolidate all of the agency's three-dimensional models, including ROM, RADM, and others. Another international initiative for designing and guiding the development of integrated, user-friendly Comprehensive Modeling Systems (CMSs) has been undertaken by the Consortium for Advanced Modeling of Region Air Quality (CAMRAQ) (28), which involves about 20 organizations from North America and Europe.

**Emission Data and Modeling**

Preparation of the emission data inputs for a photochemical modeling application, probably the most labor-intensive and uncertain/empirical component of the overall modeling process, generally consists of two steps:

- Retrieval from existing databases or estimation (using appropriate emission models) of the emission rates for each source or group of sources a) present at a specific location (major industrial point sources such as power plants, refineries, and chemical plants); b) present in an administrative area such as a county or municipality (area sources reflecting the activity of many minor residential and industrial sources); and c) corresponding to a roadway segment (traffic link or line sources). These emission rates typically represent annual or seasonal average values; they constitute a so called basic inventory.

- Processing of the information in the basic inventory to adapt it to the specific requirements of the photochemical model. This process includes:
  - adjustment of emission rates to reflect conditions specific to the ozone episode modeled and temporal (hourly) variation during the evolution of the episode;
  - spatial allocation of the emissions onto the grid of the photochemical model, using either direct information on the subcounty (submunicipality) spatial distribution of emission sources or information on the distribution of one or more appropriate surrogate factors (such as land use type or population density);
  - apportionment or speciation of emissions from each source into chemical classes. Emissions are typically reported as totals of a family of compounds (e.g., total hydrocarbons or total nonreactive organic compounds), and so they have to be partitioned into individual chemical species (actual or surrogate) that are explicitly resolved by the chemical mechanism of the model. So, total NO$_x$ must be specified as NO and NO$_2$, whereas the total reported VOC must be apportioned in classes of alkanes, alkenes, aldehydes, or aromatics. For example, when using the CBM-IV mechanism, each carbon atom of total VOC emissions is assigned to one of the following ten species: olefinic carbon bond (OLE), paraffinic carbon bonds (PAR), toluene (TOL), xylene (XYL), formaldehyde (FORM), high molecular weight aldehydes (ALD2), ethene (ETH), methanol (MEOH), ethanol (ETOH), and isoprene (ISOP). The database so developed is usually referred to as a modeling inventory.

Special types of modeling inventories are projected future baseline as well as control strategy inventories. These reflect anticipated or potential changes in inventories due to changes in levels and distribution of emission activities or due to the implementation of emission control strategies.

To organize, assess, modify, quality-ensure, and analyze the vast amounts of information contained in basic inventories, as well as to develop episode-specific inventories (for both historical and future baseline cases and for control strategy assessments), it is necessary to use specialized data management software systems. Such systems have evolved in response to
regulatory needs; the U.S. EPA has developed the Flexible Regional Emissions Data System (FREDs) for the preparation of the gridded hourly emissions, which are input to the Regional Oxidant Model (ROM). FREDs consists of a series of processing modules that, among other functions, perform the speciation, spatial, and temporal allocation of emissions. U.S. EPA has also developed and released for public usage the Emissions Preprocessing System, currently available in version 2.0 (EPS-2.0), a comprehensive system for managing the preparation of modeling inventories for UAM-IV from basis annual or seasonal county-level inventories (29). Various other modeling groups have developed additional specialized tools for managing emission inventories that can be used instead of, or in addition to, specific modules of EPS. Typically these tools represent a combined use of both standard (nonproprietary) software platforms (Fortran and C routines) and procedures developed on proprietary software packages such as SAS (for statistical analysis of data) the Advanced Visualization System (AVS; for the visualization and animation of multidimensional data sets), and the Geographical Information System (GIS) ARC/INFO (30). Visualization and GIS software is used by the GEMAP (Geocoded Emissions Modeling and Projections System), a comprehensive emission management system, which has been recently developed by Radian Corporation. GEMAP offers additional capabilities and flexibility; it includes procedures that offer alternatives to U.S. EPA's standard emission estimation models, and it can be applied to combined urban-regional scale projects with nested and variable modeling grids. GEMAP is the emission preprocessing system used in the comprehensive SARMAP and LMOS modeling studies.

**Emission Databases and Models**

**Anthropogenic Emissions.** Modeling inventories in the late 1980s and early 1990s for regional and urban applications such as the 1985 base year ROMNET anthropogenic emission inventory were based on emissions data in the 1985 National Acid Precipitation Assessment Program (NAPAP) inventory. This inventory was derived from the National Emissions Data System (NEDS), established in 1971. Recently, the Emission Inventory Branch (EIB), in a cooperative effort with the Source Receptor Analysis Branch (SRAB) of U.S. EPA's Office of Air Quality Planning and Standards (OAQPS), has developed a series of interim regional inventories (30) for the years 1987 to 1991. These inventories were based on projections from the basic 1985 NAPAP inventory and on updated emissions information for cases where such information was available. These inventories, containing county level emission estimates for the entire United States and three provinces in Canada became available through U.S. EPA's National Computer Center (NCC) in 1993, mostly for diagnostic modeling studies. Finally, addressing requirements of the 1990 CAAA, most urban areas facing ozone nonattainment problems have developed VOC and NOx inventories, typically for the year 1990, at the level of detail of and in a format compatible with U.S. EPA's Aerometric Information Retrieval System (AIRS), which has replaced NEDS (31). Information on point source emissions is maintained in the AIRS Facility Subsystem (AFS), whereas information on area sources is maintained in the AIRS Area and Mobile Subsystem (AMS) in standardized formats.

**Point sources.** Point source information in basic inventories typically includes source identification, process information, and emissions data. Source identification includes county, facility, and source codes; Standard Industrial Classification (SIC) code of the facility; and location in latitude and longitude or UTM (Universal Transverse Mercator) coordinates of each source. Process information includes Source Classification Code (SCC) or equipment codes for individual processes; stack parameters (height, diameter, gas temperature, and gas exit velocity or flowrate); control device information; operating rates and schedules; and fuel characteristics. Emissions data includes annual or seasonal estimates of VOC, NOx, and CO emissions for each process within the facility.

An important regulatory concept in the development of point source inventories is that of rule effectiveness. This is introduced to modify reported information, since experience indicates that regulatory programs are typically less than 100% effective. Accordingly, a rule-effectiveness factor is applied to emission estimates (in addition to the control factors associated with each measure) to account for less than full compliance. The current ozone policy states that a default factor of 80% can be used to estimate rule effectiveness in base year inventories. Alternatively, states are given the option of deriving local category-specific rule effectiveness factors within tightly prescribed guidelines (32).

**Area sources.** Existing inventories usually contain collective emissions estimates at the county (or municipality) level for sources considered too minor or too numerous to be handled individually in the point source inventory. In addition to small stationary sources, the basic area source inventory often includes emissions from off-highway mobile sources such as aircraft, locomotives, and off-road vehicles. It is possible that many sources, which are treated as individual point sources in the basic inventory, are aggregated during the preparation of the modeling inventory. Specific cutoff limits for representing a source either as an individual point or lumping it in an areal sum are usually established based on an evaluation of the number and distribution of sources in the domain.

**Mobile sources.** Mobile sources of emissions include moving vehicles classified as the following types: on-road vehicles (registered light duty automobiles and trucks as well as medium and heavy duty vehicles); off-road vehicles (farm equipment, construction equipment, snowmobiles, and off-road motorcycles); aircraft; railroad locomotives; and vessels.

The emission factors used to estimate emissions from on-road motor vehicles vary nonlinearly with a variety of parameters including vehicle type, vehicle speed, fuel volatility, vehicle fleet characteristics, ambient temperature, diurnal temperature variations, and vehicle fleet inspection program characteristics. Empirical models such as the MOBILE series of mobile source emission factors, available from U.S. EPA's Office of Mobile Sources (EPA OMS), are commonly employed to estimate on-road vehicle VOC, NOx, and CO emission factors. These emission factors, reported as mass of pollutant per vehicle mile travelled, are then combined with an activity level such as vehicle-miles travelled (VMT) to calculate estimates of on-road vehicle emissions. Ideally, link-specific traffic volumes and speeds should be used to generate emission estimates. Various inventory classification schemes may then be employed to aggregate these emissions into a manageable number of categories such as vehicle class or road type. Emissions for each category are typically reported as a county (or municipality) total in annual or seasonal state inventories.

To facilitate accurate spatial allocation, speciation of mobile source VOC emissions, and analysis of detailed control strategies, emissions from on-road mobile sources are reported by both vehicle type and roadway
classification. The MOBILE models distinguish nine vehicle classes based upon gross vehicle weight (GVW) and fuel consumption type (gasoline or diesel fuel). The Federal Highway Administration (FHWA) maintains statistics for 13 types of roads: rural and urban interstates, principal arterials, minor arterials, major collector, minor collector, local, and other freeways and expressways. Emission factors vary by road type because of the variation in parameters such as speed and fleet distributions associated with different road types. It should be noted that information contained in older inventories may not be so detailed; the NAPAP inventory included only four vehicle classes and three road types.

In addition to being categorized by vehicle type and road class, on-road mobile source emissions must be disaggregated in terms of component emissions (exhaust, evaporative, running loss and refueling

Biogenic Emissions. In recent years, it has been recognized that biogenic emissions (naturally occurring emissions from vegetation) can contribute significantly to the total emission inventory, even in predominantly urban regions. Some of the naturally occurring organic species are quite photochemically reactive (isoprene). Accordingly, the modeling inventory must include an estimate of biogenic emissions. Researchers at Washington State University and U.S. EPA have developed a modeling system, the Biogenic Emissions Inventory System (BEIS), for estimating hourly gridded biogenic emissions (29,32). BEIS calculates individual biogenic emission rates for chemical species such as isoprene or monoterpenes for each vegetation type as the product of leaf biomass factor, an emission factor, and an environmental factor accounting for solar radiation and leaf temperature. Total emission rates are calculated by summing emissions from different vegetation types.

The leaf biomass database used by BEIS is derived from land use data in the Oak Ridge Laboratory’s Geoecology Data Base. The land use database is resolved at the county level and includes acreages for forest types, agricultural crops, and other areas such as urban, grassland, and water. Each of the forest types in the land use database is assigned to either oak, other deciduous, or coniferous forests. The leaf biomass for each forest group is partitioned into four emission categories: high isoprene deciduous, low isoprene deciduous, nonisoprene deciduous, and coniferous. Emission factors for four hydrocarbon species (isoprene, α-pinene, monoterpenes, and unidentified) are included in BEIS, as well as estimates of NO2 emissions from grasslands.

Studies indicate that biogenic emissions from most plant species are strongly temperature dependent; isoprene emissions also vary with solar intensity. The emission factors used by BEIS are adjusted to account for these factors. BEIS also simulates the vertical variation of leaf temperature and sunlight within the forest canopy. The canopy model in BEIS assumes that sunlight decreases exponentially through the hypothetical forest canopy; the rate of attenuation depends on the assumed biomass distribution. Visible and total solar radiation are calculated for eight levels in the canopy and used to compute the leaf temperature at each level.

Temporal Resolution and Spatial Allocation of Emissions

According to U.S. EPA Guidance (32), ideally, each major emitting facility should be contacted to obtain hourly operating records for the modeling episode, or, if this information is unavailable, representative operating schedules for a typical ozone season day. Certain local agencies may also have this type of temporal information. Resource limitations, however, generally make determination of source- or episode-specific operating schedules impractical except for the largest emitters in the area. Some sources for which this type of data may be available include the following: power plants (which generally keep detailed, hourly records of fuel firing rates and power output for each day of operation), major industrial facilities such as automotive assembly plants and refineries, and tank farms. Empirical relationships are used to resolve the diurnal variation of smaller point sources and of area sources (29).

Sources must also be assigned to the appropriate cell of the modeling grid. Since point source locations are typically known to the nearest tenth of a kilometer, it is straightforward to assign them to specific grid cells. Area source emissions are typically only resolved to the county (or equivalent) level in annual inventories and thus must be disaggregated to the grid cell level. County-level area source emissions estimates can be apportioned to grid cells using either of two approaches: a) in certain cases, determining the activity levels and emissions of some area sources directly for each grid cell may be feasible; or b) more commonly, the emissions are apportioned by assuming that the distribution of the area source activity behaves similarly to some spatial surrogate indicator (32). A spatial surrogate indicator is a parameter, the distribution of which is known at a subcounty level, that is assumed to behave similarly to the activity levels of interest. Commonly used spatial surrogate indicators include land use parameters, employment in various industrial and commercial sectors, population, and dwelling units. Different surrogate indicators should be used to apportion emissions for the various area source categories depending on which of the available indicators best describes the spatial distribution of the emissions.

The U.S. Geological Survey (USGS) maintains a comprehensive computerized national database of land use distribution data based upon a standardized classification system. The USGS data files, available in both digital and character formats, contain data for many regions of the country in terms of 4-hectare grid cells (200 m × 200 m). Items contained in the database for each individual grid cell include UTM zone, UTM Easting and Northing, land use and land cover attribute code, political unit code, USGS hydrologic code, census county subdivision or SMSA tract code, federal land ownership agency code, and state land ownership code. Since a given modeling region will often contain over 500,000 four-hectare grid cells, manipulation of such large amounts of data is best accomplished with the aid of a computerized information management system such as a Geographical Information System (GIS). As part of the transportation planning process routinely performed in larger urban areas, employment and other demographic statistics are aggregated at the zonal level. These statistics can be used instead of, or in addition to, land use patterns to obtain the information needed to apportion area source emissions to the subcounty level.

It should be noted that updated information on land use and land cover can be obtained via analysis of commercially available satellite (LANDSAT) imagery. This has been done for the LMOS study.

Speciation of Emissions

Generally, the basic annual inventory will contain estimates of either total VOC or nonmethane VOC, depending on what emission factor information is used for computing emissions. The basic approach for allocating VOC into the classes needed by a photochemical model is to employ a set of split factors that distribute a certain fraction of the VOC total into each class. Ideally, VOC split factors should be source-specific, reflecting the actual composition of VOC emissions from each
individual source. Resource limitations and unavailability of detailed composition data for certain VOC mixtures such as solvents often render the compilation of source-specific speciation data impractical, except maybe for a very few large emitters. An alternative is to use generalized VOC speciation data from the literature to develop VOC split factors by source type. To develop CBM-IV split factors from literature speciation data, the individual chemical compounds typically present in the emissions from each source type and their weight fractions in the emissions mixture must first be identified. Then, each of the chemical compounds present in the modeling inventory must be classified according to the CBM-IV mechanism or whatever mechanism is employed by the PAQSM used. The U.S. EPA (32) has identified over 250 emission profiles for various point and area source categories. Split factors for CBM-based modeling are expressed in units of moles of carbon bond species per gram of total VOC and represent a weighted composite of the carbon bond class assignments for each of the chemical compounds present in the mixture. The accurate speciation of emissions is a tenuous and error-prone process that is complicated by the fact that the source categories and subcategories chosen for the basic inventory may fail to distinguish between sources having substantially different emission compositions, thus requiring different sets of split factors.

Emissions Projections for Future Years. According to U.S. EPA Guidance (32), the recommended approach for projecting emissions from major point sources is to obtain information on each facility by contacting the plants directly or through questionnaire. Permit applications submitted to various federal, state, and local agencies should also be screened to get information on expected expansion or new construction. Local Metropolitan Planning Organization (MPOs) and other planning bodies may also have information on projected industrial expansion and can evaluate the reasonableness of plans submitted by regulated industry. Usually, projection information is not available for every facility in an area of interest. Furthermore, many facilities in certain source categories will be too small and too numerous to justify the collection of projection information individually. A reasonable approach to projecting growth and emissions is to evaluate the growth trends for the facilities for which projections are known and to apply them to the facilities for which no information is available. In other cases, the rate of growth of activity may be assumed equivalent to that of some growth indicator category for which projections are available. Sources of growth indicator projections include local MPOs and the U.S. Department of Commerce’s Bureau of Economic Analysis (BEA). BEA makes projections at the state and Metropolitan Statistical Area level for a subset of the two-digit SIC designations associated with each emission source category. The BEA projections of industrial employment are regularly updated and are used as default alternatives in the absence of local projections as general indicators of growth.

It should be noted that, just as the quantity of emissions may change in an area from the base year to any projection year, the composition of these emissions may change as well. So, different VOC and NOx split factors may need to be used for each projection year, at least for important sources for which such projected compositional changes can be estimated. A major source for which this is an important consideration is motor vehicles; changes in emissions control technology and use of alternative or reformulated fuels are expected to result in significantly different VOC split factors in projection years. In general, the same emissions factors are used to estimate biogenic emissions for both base and projection years. However, one may wish to incorporate the effects of anticipated changes in land use patterns on spatial allocation of biogenic emissions into the projection inventories if this type of data is available.

Special effort is needed to establish control strategy projections; these are estimates of emissions for some future year that consider the effect of proposed control measures. Control strategy projections should be made for the same years as the future baseline projections to facilitate comparison of the relative effects of each strategy, as well as to determine which strategy provides the necessary control of ozone precursor emissions (32).

**Meteorological Data and Modeling**

**Meteorological Databases**

There are three major types of sources for surface meteorological observations: Class A and B National Weather Service (NWS) Stations, National Data Buoy Center (NDBC) moored buoy and coastal land stations, and meteorological instruments located at many air quality stations that report to U.S. EPA’s Aerometric Information Retrieval System (AIRS). In addition, there may be meteorological information available that is collected at other locations (power plants, educational facilities, etc.). Typically, surface-weather stations report wind speed and direction, air temperature and dew point temperature, atmospheric pressure, and cloud amounts and heights. Most NWS stations are located at airports and report measurements on an hourly basis; however, some stations at smaller airports only make measurements during the day and evening. The NWS data are not hourly averaged data: typically they are instantaneous or short-term average measurements taken at 5 to 10 min before each hour. For coastal regions, NDBC stations are also important; they include stations on moored buoys, deep sea buoys and Coastal-Marine Automated Network (C-MAN) stations which are located at lighthouses, beach areas, and offshore platforms; they also report hourly measurements. Some of the air quality sites that monitor surface meteorological parameters provide continuous records that can be used for deriving hourly-averaged values rather than assuming an instantaneous wind velocity to persist unchanged over the next hour.

Upper air meteorological measurements are made on a routine basis at a very limited number of NWS stations. Rawinsondes are launched twice daily by the NWS at 0000 UTC (1900 EST) and 1200 UTC (0700 EST). Upper air data are reported at significant levels where large changes in temperature, humidity, or winds occur, as well as at mandatory levels (i.e., at 850, 700, 500, 300, and 200 mb atmospheric pressure) throughout the troposphere and lower stratosphere. The vertical resolution varies and is typically coarse (about 100 to 200 m).

**Meteorological Models for Windfields and Mixing Heights**

Meteorological preprocessors for PAQSMs range from schemes that simply interpolate observed surface-level wind data to complex models based on the fundamental equations of atmospheric flow. A typical classification of approaches in use is:

- **Objective analysis procedures.** Values of the wind field at required points are obtained by a weighted interpolation of observed data.
- **Diagnostic procedures.** Diagnostic methods use some or all of the governing equations to solve for the wind field, with the assumption that a steady-state solution adequately represents meteorological conditions for the short period of interest.
Prognostic methods. These methods are based on the numerical solution of the coupled, turbulent conservation equations for mass, momentum, energy, and water vapor together with the appropriate thermodynamic state equations. These are the so-called primitive equations. Existing formulations employ either first or higher order turbulence closure schemes and result in complex, computationally intensive models.

The choice of technique depends primarily upon the spatial and temporal representativeness of the available observational data. The simplest method, objective analysis, involves no physics but only interpolation and extrapolation of the available data. Constraints may be placed on the field, i.e., the requirement that the vertical velocity at the top of the modeling domain must be zero so that no material can escape from or intrude across the upper boundary. The primary advantage of objective analysis is that it is computationally inexpensive. Disadvantages arise because available observations are often unrepresentative of air flow in certain portions of the domain (this is especially true in regions of complex terrain) and are inadequate in number or spatial coverage. These available observations also do not provide a sound basis for extrapolation at the boundaries of the region. For complex terrain or coastal environments, it is tenuous to interpolate between and extrapolate from surface observational sites except with an unusually dense monitoring network. In most cases, the routinely available rawinsonde network sounding data are even more severely limited due to the large distances between sites and because soundings are made only every 12 hr. Even the impressive array of instrumentation deployed in the LMOS and SARMAP field programs cannot provide enough observations to resolve all of the important locally forced flows that contribute to the circulation and mixing of primary and secondary pollutants. It is generally agreed today that objective analysis methods fail to produce self-consistent atmospheric fields with the accuracy required by regulatory PAQSMs.

The simplest diagnostic methods are based on satisfying only the conservation of mass equation. These methods impose mass consistency on the three-dimensional wind field subject to constraints such as vertical velocities not exceeding a certain limit. An initially assumed wind field, frequently generated by an objective analysis method, is iterated upon so that mass is conserved. Diagnostic wind modeling may involve relatively simple estimation of complex terrain effects such as the deflection and blocking of air flow by complex terrain and, in certain models, an estimate of upslope and downslope flow through the heating and cooling of slopes. These estimates are usually combined with the objective analysis of observations. With a diagnostic wind model, fewer observations may be required than with simple objective analysis to produce a wind field. The main disadvantage is still that diagnostic wind models cannot generate certain air flow features, such as the sea breeze, that are important in air quality simulation unless these features are well represented by surface and aloft observations. Often the vertical velocities produced by a diagnostic model are unrealistic, and in regions of complex terrain, local horizontal flow velocities may often be an order of magnitude too high (11). Since diagnostic models are not based on time-dependent balance equations, there is no inherent dynamic consistency in the winds from one hour to the next. So, the calculation of the flow field at a given hour is not influenced by the calculation for the previous hour. This can be a serious problem in situations involving secondary circulations such as land-sea or land-lake breezes that take several hours to develop and whose three-dimensional character is poorly characterized by even the most intensive sampling networks.

Prognostic or primitive equation models are based on numerical solution of the coupled, nonlinear, mass, momentum and energy balance equations of the atmosphere. Starting from a set of initial conditions representing the large-scale flow, the model simulates the response of the air flow within the domain of interest to differential heating of the surface. A prognostic model is intended to represent all relevant physical processes that are occurring within the model domain on the scales of interest. Because they explicitly address the various physical processes governing atmospheric flows, they have the potential for describing a number of wind regimes that are relevant to air pollution modeling such as flow reversal, daytime upslope flows, wind shear, and mesoscale thermally induced circulations. Another major advantage is that, in addition to the mean wind field, prognostic models simulate the temperature field, from which one can determine the mixing height and the stability characteristics of the atmosphere, both of which are required inputs to photochemical air quality models. Prognostic models represent the state of science in meteorological modeling. Comprehensive programs (e.g., LMOS and SARMAP) are currently using them to explore the extent to which the prognostic approach, when used in application-oriented studies, provides improved wind fields compared with older methods.

Drawbacks of prognostic models include the need to gather detailed data for model performance testing and the large computational costs. Indeed, prognostic models may require as much or more computer time than PAQSMs. Another disadvantage is that prognostic models do not necessarily reproduce available observations, since they do not rely on measurements after their initialization. Numerical approximations, physical parameterizations, and initialization problems are among the potential sources of error growth in model forecasts that can cause model solutions to deviate from actual atmospheric behavior. Various methods have been developed to mitigate such problems; they include post-processing, dynamic initialization, and Four-Dimensional Data Assimilation (FDDA) techniques.

Postprocessing refers to methods where output fields from prognostic models are selectively adjusted through a series of objective techniques to improve the realism of the resultant fields. A prognostic model that is initialized with observed three-dimensional fields of wind, temperature, and moisture can generate non-meteorological waves when these initial conditions do not contain a dynamic balance consistent with the model formulation. A dynamic initialization procedure can be used to bring these initial conditions into dynamic balance, i.e., consistency with the governing equations so that the model can integrate forward with a minimum of noise and a maximum of accuracy. This is based on a presimulation integration of the model equations to produce a set of dynamically balanced initial conditions. FDDA encompasses a class of procedures in which observational data are used in conjunction with prognostic models to improve the estimates of the latter. The most common use of FDDA is known as Newtonian relaxation, or simply as nudging, where model estimates at a particular time interval are relaxed toward the observations by adding artificial tendency terms to the governing prognostic equations. As an example, a linear term is added to the momentum equations to nudge the dynamic calculation toward the observed state at each time step, in regions where data are available. FDDA is finding increasing application in wind field generation for photochemical modeling applications (1,11).
**Wind Field Modeling in Regulatory Applications**

**ROM Wind Fields.** The meteorological field generators for ROM have been designed to make use of the routine NWS surface and upper air information. These data, together with topography and land use data, are processed to generate input files to the core ROM2.2 model. In the early stages of processing, the surface and rawinsonde data are interpolated and averaged to generate intermediate level fields such as gridded surface fields of temperature, cloud cover, and solar zenith angle and vertical profiles to meteorological parameters at prescribed levels (e.g., 25-mb increments, 50 meter increments). A number of meteorological parameters, including horizontal eddy diffusivities, are also derived from the basic measurements. In the later stages of processing, three-dimensional, time-dependent meteorological fields are prepared for input to the core model. The gridded hourly surface meteorological parameters developed by the ROM processors include the Monin-Obukhov length, surface heat flux, friction velocity, surface temperature, surface relative humidity, surface wind speed, fraction of sky covered by cumulus clouds, cumulus cloud-top heights, wind fields in the cold layer (i.e., the nocturnal layer beneath an inversion), atmospheric density, solar zenith angle, water vapor concentration, eddy diffusivities, and effective deposition velocities.

The procedures for constructing ROM2.2 wind fields are based on combined diagnostic analysis and simplified prognostic modeling. The approach used interpolates observed surface and twice-daily upper level wind measurements that have been adjusted statistically to account for measurement errors and the variability between point measurements and regional scale wind patterns. The upper air data are linearly interpolated in time to produce hourly profiles at 25-mb levels. Assuming a shallow, two-dimensional fluid, mass balance is applied in conjunction with three physical constraints: wind fields constructed at monitoring sites are forced to match observations as closely as possible; in matching the observations, the algorithm attempts to minimize total kinetic energy; and the solution is constrained to fit similarity law describing the kinetic energy distribution over the various scales of motion pertinent to regional modeling. At night, when a surface inversion covers most of the modeling domain, a simple prognostic model computes the bottom layer winds rather than relying on observed data.

**UAM Wind Fields.** The current release of EPA's UAM-IV includes the Diagnostic Wind Model (DWM) (33) as the recommended wind field generator for this urban-scale photochemical model. The DWM is a hybrid objective/diagnostic model that follows a two-step procedure. In step 1 a domain-scale wind, consisting of a single horizontal wind vector for each elevation, is estimated from available surface and upper air synoptic data. The domain-scale wind is subsequently adjusted for the kinematic effects of terrain such as lifting, blocking, and flow acceleration. Thermodynamically generated influences such as mountain–valley winds are parameterized. So, this step finally produces a horizontally varying wind field for each layer within the DWM modeling domain. Typically, 10 to 12 horizontal layers are used in the vertical direction. Next, in step 2, available hourly surface and upper air measurements are objectively combined with the step 1 hourly diagnostic flow fields to produce a resultant wind field that matches the observations at the monitoring points and obeys the general constraints of topography in regions where data are absent. DWM contains a number of user-specified options whereby different final flow fields may be produced, depending upon selection of various smoothing and weighing parameters. The final output of DWM is a set of hourly-averaged horizontal wind fields for each model layer. Once the winds are created by DWM, they must be mapped onto the grid of the photochemical model. This function is accomplished in a two-step process: DWM winds are assigned to the photochemical model grid using simple linear interpolation, and the three-dimensional wind divergence is computed in each grid cell, and an iterative scheme is used to minimize this divergence to a user-specified level. The output of this process consists of almost nondivergent horizontal wind components for input to the photochemical model.

**Prognostic Models in Regulatory Applications.** Prognostic mesoscale meteorological models have been or are currently being used in support of urban and regional ozone regulatory modeling applications across the United States, including the LOMOS study, the SARMAP study, Houston, Beaumont–Port Arthur, Sacramento, San Francisco, Ventura–Santa Barbara, the Los Angeles Basin, San Diego, and the New Jersey–Philadelphia modeling domains. Most of these studies have endorsed one of two models that are considered to represent the present state of science in applications-oriented prognostic modeling (34). These are the Mesoscale Model Versions 4 (hydrostatic) and 5 (nonhydrostatic) (MM4/MM5), developed by Penn State University and the National Center for Atmospheric Research (NCAR), and the Coast and Lake Regional Atmospheric Modeling System (CAL-RAMS), a public domain, nonhydrostatic model, that has evolved from the Colorado State University Mesoscale Model (CSU-MM). MM4/MM5 and CAL-RAMS have many overlapping attributes including applicability to a variety of spatial scales and ability to incorporate multiple nested grids; their capabilities have continued to expand during recent years. The differences are currently rather limited and often deal with the emphasis of applications. Descriptions of these models are beyond the scope of this discussion; summaries and discussions of their applications can be found in Georgopoulos, Reynolds et al., Pielke and Lyons (11,34,35). According to Reynolds et al. (11), MM4/MM5 holds a slight advantage over CAL-RAMS in modeling situations involving large regional scales (20–40 km grid resolutions or larger), lengthy episodes (on the order of a week or more), and in situations where data assimilation is vital to the success of the modeling because it has been applied extensively with FDDA. In contrast, for episodes of limited duration, particularly in situations where the relevant grid scale is of order 1 to 5 km (associated with intense thermodynamically driven circulations), the CAL-RAMS model might be the preferred choice. Data assimilation is also increasingly being used with various versions of CAL-RAMS (11).

Regarding recent comprehensive modeling and field studies, CAL-RAMS has been used to develop meteorological fields for ROM2.2 and for UAM-V for LOMOS while the MM4 model was chosen as the prognostic meteorological model for the SARMAP study.

It should also be mentioned that the plan for the development of U.S. EPA'sMODELS-3 comprehensive modeling system also calls for meteorological inputs to be supplied by prognostic models (27). The MM4 model is presently being examined by U.S. EPA for this purpose. However, consistent with the emphasis on modularity in MODELS-3, other models will be incorporated in it as well.

**Model Evaluation: Operational and Diagnostic**

There exists a pressing challenge today to use valid criteria and methods for
determining whether a photochemical model performs well enough for use in regulatory decision making. In 1988-1990, the California Air Resources Board (CARB) sponsored a study to establish the basis for consistent photochemical grid model performance evaluations in the near term and to provide a framework for performance evaluation research over the longer term. Many of the conclusions and recommendations of that study (10,36) were endorsed by scientific panels, the U.S. EPA, and various state environmental agencies (2-4,37) and have been incorporated in procedures now employed in ozone attainment SIP modeling studies (29). In the following, we will summarize the general recommendations of the CARB study in the context of model performance evaluation for regulatory applications. A distinction between operational and diagnostic evaluation corresponding to two levels of analysis of the modeling process was introduced, and relevant evaluation procedures were recommended for each of these levels. Statistical and graphical performance evaluation measures, as well as standardized investigative simulations, were suggested for operational use. Diagnostic model evaluation methods are recommended to develop greater insight into the strengths and weaknesses of a particular model and the associated databases than is provided by routine operational procedures. In complex modeling situations, or when the operational performance evaluation results are suspect, these diagnostic procedures should be an essential component of the overall evaluation process.

**Operational Evaluation Procedures**

Several statistical (numerical) and graphical procedures can be used for assessing the performance of grid-based PAQSMs. Recommended methods include the calculation of peak estimation accuracy indices, statistics based on concentration residuals (i.e., the deviations of estimated from observed values) and time series of estimated and observed hourly concentrations. Graphical representations can complement the numerical measures, providing additional insight into model performance. Indeed, certain features of a PAQSM are best analyzed through graphical displays that can provide information such as the relationship between the various measures of peak estimation accuracy, the temporal correlation between estimates and observations, the spatial distribution of estimated concentration fields, the correlation between hourly pairs of estimates observations and residuals to 1990, the variation in bias and error estimates as functions of time and space, and the degree of mismatch between model estimates and point measurements.

Recommended operational model performance evaluation measures involving various types of comparisons between hourly model estimates and observations, and the associated investigative simulations are summarized in Figure 5; an explanation and discussion of these measures is presented in the Appendix. It should be mentioned that, while these measures in principle may be applied to any primary or secondary pollutant, in practice due to data limitations, their use is intended primarily for O₃ and NOₓ concentrations.

The CARB study did not endorse rigid criteria for model acceptance or rejection. Instead, based on review of over 15 years of model development and testing studies, it was recognized that photochemical grid models generally produce peak (unpaired) estimation accuracy, overall bias, and gross error statistics in the approximate ranges of 15 to 20%, 5 to 15%, and 30 to 35%, respectively, when calculations are compared with observations. For model simulations falling within these ranges, some additional diagnostic analyses may still be appropriate to establish adequate understanding of model response, but, unless some type of inordinate behavior is observed in these analyses, the results should generally be considered acceptable. For model results outside any one of these general ranges, it should be incumbent on the modelers to explain why the performance is poorer than that commonly achieved in similar

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**Figure 5.** Recommended operational performance evaluation measures and simulations for the regulatory application of Photochemical Air Quality Simulation Models.
applications. The modelers should also explain whether the causes of poorer performance will adversely affect the use of the model in control strategy evaluations. This methodology provides reviewing regulatory agencies and policy makers with a general performance target, but still guards against the inappropriate rejection of less accurate model simulations when appropriate and explainable reasons can be provided.

**Diagnostic Evaluation Procedures**

Recommended diagnostic evaluation analyses (10) include graphical analyses of concentration residuals, multi species comparisons, mass fluxes and budget calculations, and various sensitivity-uncertainty studies, as summarized in Figure 6.

**Analysis of Residuals.** The set of deviations between estimated and observed concentrations in a PAQSM performance evaluation contains significant, though lumped, information about contributions to errors associated with: a) the air quality data used for comparison with model output, b) the soundness of the model formulation, and c) the adequacy of the data supplied as input to the model. In addition to the operational evaluation procedures that are based on the analysis of concentration residuals, insight into model performance can be gained by plotting these residuals against selected variables in order to identify patterns of aberrant behavior. If correlations (relationships) between the residuals and one or more selected variables can be found, the emergent patterns may be suggestive of the causes of failure or inadequacy in the model. Variables that may be selected for plotting against residuals include time, geographical location, concentration levels, meteorological variables, emissions, and deposition rates. Plots can be made for the full region of interest and for the full duration of the simulation, or for subregions, selected time periods, and specified ranges in variables.

**Multispecies Comparisons.** The use of evaluation procedures that test photochemical model performance for species other than ozone is strongly recommended. Multispecies comparisons provide a more robust basis for accepting or rejecting a model (or a model simulation); they significantly improve the chances that a flawed model will be identified. Adequate model performance for several reactive species increases the decision maker's assurance that correct ozone estimates are not a result of chance or fortuitous cancellation of errors introduced by various assumptions. In addition to ozone, the following species should be subjected to the performance evaluation if suitable data are available: NO, NO₂, total and speciated VOCs, H₂O₂, HCHO, PAN, HNO₃, and particulate nitrate.

**Mass Fluxes and Budgets.** Four types of mass balance and flux calculations are recommended for a detailed performance evaluation: a) calculation of mass fluxes into and out of domain boundaries; b) calculation of mass fluxes into and out of the mixed layer; c) calculation of surface deposition fluxes; and, d) reconciliation of emission, transport, transformation and removal terms in a closed budget.

Mass balances and flux calculations have been performed to a limited extent in the past, and little guidance can be offered with respect to how these results should be judged. The ultimate value of these calculations for diagnostic performance evaluation and stress testing will evolve as more experience is gained in their use and interpretation.

**Sensitivity–Uncertainty Analysis.** Sensitivity analysis is an essential component of model performance evaluation and should be conducted as part of any comprehensive modeling study. Sensitivity analyses help to reveal internal inconsistencies in the model, identify the inputs that dominate the model's operation, and support analyses of error propagation through the model. Their results help develop guidance for model refinement and data collection programs (10,25). For a sensitivity analysis, key input variables and parameters need to

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**Figure 6.** Recommended diagnostic evaluation tests and analyses for the regulatory application of Photochemical Air Quality Simulation Models.

[1] MV: Meteorological variable category (e.g., wind, mixing height, etc.)
[2] ER: Emission rate category (e.g., motor vehicles, biogenics, etc.)
[3] SDR: Surface deposition rate
be identified and their levels of uncertainty estimated. These uncertainty bounds are then propagated through the model, either singly or in concert, to provide estimates of the uncertainties in the estimates. In an ideal case, estimates of probability distributions associated with the values of selected inputs (probability encoding) would be determined, and the corresponding probability distributions of model outputs would then be calculated via a repetitive application of the model employing an efficient algorithm for random input sampling. In practice, such an approach is presently not feasible for comprehensive PAQSMs, although it has been applied to individual model components (e.g., the chemistry module). Of particular value is a qualitative analysis of the results of the sensitivity runs from the viewpoint of the response of the model expected from its underlying physics and chemistry.

Discussion and Research Needs

In the following, research needs in the field of ozone modeling are identified in three broad categories: model evaluation and refinement, uncertainty characterization, and regulatory application in an ozone attainment framework.

Research Needs in PAQSM Evaluation and Refinement

Testing Model Response to Emission Changes. The adequacy of a photochemical model in correctly estimating the effects of emission changes on ambient air quality should be evaluated directly by examining model performance for applications involving significantly altered emission strengths and spatial patterns. Ideally, such a performance evaluation should be performed with emission inventories for the same region that correspond to years sufficiently separate in time. If such a procedure is not feasible, then, as an alternative, one should evaluate the model for two or more different regions, using input data of comparable quality.

Emission Models. Preparation of emission inputs is probably the most uncertain modeling step involved in a regulatory PAQSM application. Improving emission inventories is of considerable importance, since any control strategy decisions made, with or without the aid of models, are only as good as the emissions estimates upon which they are based. Directions in the evaluation and refinement of emissions estimates can include the following:

- Top-down versus Bottom-up Calculations. Emissions estimates should be developed, where possible, by estimating emissions of individual sources or groups of sources and then aggregating them and, independently, by calculating gross or integrated emissions rates. Comparison of the different estimates could provide insight into potential sources of error in modeling inventories.
- Mass Balance Calculations. Mass balance calculations need to be considered as part of the emissions modeling process. For example, nitrogen balances can be made for determining emissions estimates and uncertainties for animal wastes, chemical fertilizer use, and wastewater treatment plant emissions.
- Comparison of Ambient Air Ratios versus Emissions Ratios. One should compare ratios of ambient concentrations of selected pollutant species measured near the source with estimated ratios of emissions rates of the two species. Analysis of differences in ratios may be used in estimating the magnitudes of emissions uncertainties. Furthermore, ambient VOC/NOx ratios and their trends and evolution must be continuously monitored and analyzed. With the imposition of VOC controls, VOC/NOx ratios in many regions may be decreasing, suggesting that NOx controls may provide a relatively less effective means of reducing O3 than further VOC controls.
- Indirect Confirmations. External information should be used to the fullest extent possible to corroborate direct emissions estimates. For example, an energy balance on fuel consumption statistics has been used in estimating sulfur emission rates.
- Source Testing. Emissions of selected sources should be determined through direct measurement. Because of the large attendant costs, one must develop specific guidelines for establishing this need and procedures for cost-benefit assessment.
- Improved Motor Vehicle Emissions Estimates. There is a need to improve the characterization of the driving cycle, including the degree of representativeness, extent of variability due to changes in commute characteristics, driver characteristics, and other influences. It should be noted that quality CO measurements have been useful for helping to identify possible problems in motor vehicle emissions estimates.
- Improved Biogenic Emissions Estimates. Methods for evaluating the relative contributions of biogenic and anthropogenic emissions to precursor levels in each nonattainment area must be developed. Detailed VOC composition analyses or other tracers of opportunity may provide a means to better establish the importance of biogenic emissions and hence the expected effectiveness of VOC controls.
- Specially Designed Field Studies. Field studies should be considered specifically for emissions determination. Examples include the SCAQS tunnel study for corroborating estimates of emissions from motor vehicles.

Meteorological Models. Meteorological preprocessors for PAQSMs, of both the diagnostic and the prognostic type, should be evaluated independently for their ability to reproduce observed atmospheric property fields (wind speeds and directions, mixing heights, temperatures, etc.) in a wide range of situations. Various numerical and graphical measures for quantifying and analyzing the performance of meteorological models are presented and discussed in detail in Tesche et al. (10). Although ongoing intensive field studies will provide some databases for such evaluations, it is necessary to plan studies for additional regions in the United States and also to consider enhancements to routinely operating meteorological monitoring networks. Clearly, a denser network of upper air stations is required to produce reliable mixing height fields and to resolve and diagnose phenomena such as low level jets, and terrain channeling that affect fluxes of O3 and precursors into and out of major source regions. In fact, meteorological data aloft are needed on an hourly rather than twice-daily basis to evaluate model performance and to ensure that models represent the most important processes contributing to O3 exceedances. Data on hourly upper air winds and temperature can be obtained with radar profilers and RASS (radio acoustic sounding systems).

Coastal surface meteorological sites are needed to identify conditions associated with onshore/offshore flow and land–sea breezes. If flow offshore is important, then these phenomena must be represented adequately by regulatory models and the associated databases.

Chemistry Models. The gas-phase chemistry submodel is one of the few components of photochemical models that can be and has long been independently evaluated. Multispecies mechanism testing with environmental chamber data is the best available method of evaluating the chemistry for PAQSMs. The recommended procedure
for mechanism testing \((10)\) follows a hierarchical approach where testing is initiated at the lowest level in the hierarchy (NO-air experiments to test the inorganic reactions and the chamber characterization procedures) and proceeds to the highest level (longer carbon chain hydrocarbons by stepwise addition of species with increasingly complex chemistry.

To better evaluate and refine the chemistry components of PAQSMs, research can focus on the following areas: acquiring additional smog chamber data from existing facilities and with existing methods; developing and applying methods to collect data for species not presently measured in existing facilities; and developing cleaner environmental chambers. Data collection and methods development efforts should focus on the identification and subsequent reactions of aromatic ring-fragmentation products, the radical yields in ozone-olefin reactions, the oxidation mechanisms of alkanes with more than five carbon atoms, acquisition of photolytic data for carbonyls, and considerations of multi-day transformations.

In the future, the chemistry of regulatory ozone models should be enhanced to include multiple phases. Some of the regional application-oriented models already incorporate multiphase phenomena, since they were developed with focus on acid precipitation. It is generally accepted today that the inorganic nitrate estimates from these models are fairly inaccurate because they lump together two species that deposit at very different rates (i.e., nitric acid deposits very rapidly while aerosol nitrate deposits slowly) \((10)\). Improved techniques are available for measuring nitric acid and aerosol nitrate as well as peroxy acetyl nitrate (PAN) in the field. Modifying existing models to incorporate the nitric acid–aerosol nitrate equilibrium chemistry could make it possible to evaluate the nitric acid and aerosol nitrate estimates as part of multispecies comparisons that would be useful from the perspective of regulatory ozone model evaluation. Also, one could compare the observed and estimated ratios of PAN to total nitrate and total inorganic nitrate (TIN) to total nitrate to assess possible biases in the VOC and NO\(_x\) inputs to the models.

**Deposition Models.** Photochemical grid models include transport algorithms and chemical reaction resistance calculations to estimate dry deposition rates as functions of time and location. Evaluations of this methodology are needed to ensure that the procedures used agree with currently accepted boundary layer theory and surface resistance experiments. Tesche et al. \((10)\) provide specific recommendations for deposition model testing.

**Sub-grid Effects and the Incommensurability Problem.** Photochemical grid models estimate average concentrations in each grid cell whereas measurements are made at a point. These different spatial and temporal scales of measurements and model estimates give rise to the so-called incommensurability problem. Another problem related to the discretized representation of the physical airshed by the numerical modeling grid is the instantaneous mixing of emissions from a source with ambient air throughout the grid cell into which the emissions are injected. Clearly there is a need to better quantify effects that are averaged or homogenized within the cell, and thus are not resolved, and to understand how spatial averaging affects modeling results. Database and modeling refinements to address subgrid effects and the incommensurability problem can be pursued in the following areas:

- **Sub-grid Scale Modeling.** There are various ways to incorporate into grid-based models certain attributes of the dynamic processes, such as chemical reaction, mixing, dispersion, and deposition, that occur at spatial scales less than the size of the modeling grid cell. Plume-in-grid models that are integrated with airshed models represent one promising direction; other concepts from non-ideal reactor modeling methods could also prove useful in this area.

- **Finer Grid Resolution.** One may reduce the dimensions of a grid cell size, although this leads to increased computing requirements. However, it should be realized that the maximum resolution of an air-shed grid depends on the method that is employed by the model to represent transport properties. For example, in the present generation of PAQSMs that use K-theory to treat turbulent transport, reduction of the cell dimension below 2 km should be expected to violate the theoretical formulation of the model under most circumstances \((23,38)\).

- **Remote Measurements.** Multiple measurements employing remote sensing techniques may permit local concentrations to be compared directly with grid cell-averaged model estimates. Equipment potentially useful for this purpose is still in development.

- **Multiple Measurements Within a Cell.** Monitoring at multiple sites within a grid cell provides an alternate means for estimating grid-averaged concentrations, although current in situ measurement costs make this approach impractical except for research purposes.

No widely applicable approach for resolving the incommensurability problem is now available. The development of economical remote measurement systems and low cost surface monitoring devices may provide routes to its resolution.

**Air Quality Databases.** There is a need to improve the routine air quality databases to provide information appropriate for PAQSM performance evaluation; the establishment of the PAMS network \((12)\) represents an important step toward this objective. Air quality data are primarily collected at the surface; thus, there is little information to assess how well the models are treating the transport and chemical transformations of \(O_3\) and precursors aloft. The offshore/onshore transport of \(O_3\) and the influence of the land–sea breeze on its formation and transport cannot be identified or quantified without shoreline measurements. Few rural air quality stations exist at upwind boundaries to quantify boundary conditions or between major source regions for measuring the interregional transport of \(O_3\). A mixture of urban and rural sites is needed to measure the full range of \(O_3\) exceedances and to evaluate model performance in areas without fresh emissions. Data on NO/NO\(_x\) are also needed in rural areas both upwind and between the major urban areas and along the shoreline. Monitors with high sensitivity (about 1 ppb detection limit) are needed in these areas because concentrations are low. There is a need to analyze colocated NO, NO\(_2\), \(O_3\), and VOC measurements to determine the extent to which \(O_3\) formation may be VOC or NO\(_x\) limited. Furthermore, simplified techniques that have been proposed for the parameterization of \(O_3\) formation prediction \((39)\) should be evaluated via comprehensive modeling.

No up-to-date databases exist for aloft air quality. This is a major problem for both model development and evaluation efforts, since there is no way to check model performance above the surface or to evaluate the adequacy of boundary conditions. In addition, without aloft air quality data, model predictions of aloft transport by a low level jet or land–sea breeze cannot be verified against air quality data, nor can model predictions of fluxes of \(O_3\) and precursors into the region or a subregion be verified. Monitoring of air quality at various above-ground levels should eventually become a permanent feature of operating networks. In addition to the enhancement of continuously operating networks, there is
the need to conduct additional comprehensive field measurement studies to support the regulatory application of photochemical models; such a study would be particularly useful for the Northeast Ozone Transport Region (NOTR). An intensive study should characterize the relative importance of local urban emissions and transport of \( \text{O}_3 \) and precursors from upwind source areas. Such information would help to establish the credibility of photochemical modeling results. Instrumented aircraft should be employed to establish \( \text{O}_3 \) and precursor levels on flux planes upwind of each urban area; vertical wind soundings on each plane would also be needed to support the flux calculations.

### Research Needs in Uncertainty Quantification for PAQSMS

The application of PAQSMSs and the interpretation of their results, including model evaluation, is complicated by the presence of significant uncertainties that are due to both the inherently random nature of atmospheric systems and errors or incomplete knowledge associated with both model formulation and model inputs and parameters.

Photochemical air pollution systems are inherently stochastic due to the nature of the atmosphere per se, as well as to unavoidable unpredictability (randomness) and incomplete knowledge regarding human activities that result in anthropogenic emissions of primary pollutants. Incomplete knowledge and randomly varying factors are also associated with biogenic emissions. This picture of the physical system is further complicated due to the following facts:

- Many transformation processes in photochemical pollution systems are strongly nonlinear, thus complicating the system response to changes in inputs.
- There is a coupling between the stochastic attributes of atmospheric transport and mixing and the nonlinearity of atmospheric chemistry. This coupling leads to subgrid turbulent kinetic processes (40) and an associated closure problem, reflecting natural uncertainty in the evolution of chemistry, in addition to that of transport.
- Physical and chemical processes take place and interact in a variety of ways over a very wide range of temporal and spatial scales. For example, chemical reaction rates range from very fast to very slow. Fast reactions have a direct impact on the locality of the emissions and can be strongly affected or even be limited by atmospheric mixing. On the other hand, slow reactions are relatively insensitive to local mixing and affect a wider, regional or global, spatial area. Treating phenomena that occur over very different scales complicates the algorithmic and numerical formulation of PAQSMSs and introduces various approximations and errors.

- Atmospheric transport and mixing processes are, in general, nonlocal and can only be approximated by local models such as gradient transport, with the approximation introducing an uncertainty in modeling. However, as mentioned in earlier sections, modeling approximations reflecting the nonlocal character of atmospheric transport and mixing, (e.g., the transient turbulence model and the asymmetric convective modeling) are currently being accepted in the formulation of air quality models.

All the above facts complicate efforts to mathematically model photochemical pollution systems and raise various problems, mostly in relation to the averaging processes involved in modeling. Ensemble averaging is required in PAQSMSs to derive deterministic equations governing the expected values of ambient concentrations.

Spatial and temporal averaging procedures are subsequently introduced in the algorithmic formulation and numerical implementation of PAQSMSs. So, atmospheric concentrations in such models represent spatial averages over a cell of the computational grid, as well as temporal averages over an appropriate computational time step. The nonlinearity of photochemical systems introduces errors and therefore modeling uncertainties through these averaging procedures. Furthermore, subgrid (subcell) variation is not revealed or taken into account in the calculations.

Uncertainty in a photochemical modeling application is associated with:

- the air pollution system itself (stochastic atmosphere, unpredictable emission-related activities); this is the natural uncertainty,
- our incomplete quantitative knowledge of information on the system, which is used either to simply execute or to evaluate the model (measurements and estimates of emissions and aerometric data): this is the uncertainty in data,
- the photochemical model itself (mathematical and computational formulation); this is the model uncertainty.

Natural uncertainty is inherent or irreducible, whereas data and model uncertainty contain both reducible and irreducible components. The irreducible uncertainty in data and models is generally a result of the presence of natural uncertainty. Reducible uncertainty can be lowered by better inventorying methods, improved instrumentation, improvements in model formulation, etc. Nevertheless, it must be made clear that the distinction between reducible and irreducible model and data uncertainties is, to a great extent, a matter of convention, since it may not be feasible to eliminate the presence of an error (reducible uncertainty) in measurement or modeling beyond a certain level. Furthermore, what is perceived as irreducible natural uncertainty may be quantified in a statistical sense, and via mechanistic modeling, better than artificial reducible uncertainty. Modeling uncertainty reflects the current model formulation and may actually change when improved theories describing the phenomena under consideration become available. Also, the averaging processes involved in model formulation unavoidably lump together natural and modeling uncertainty, and only a quantification of this lumped uncertainty may be possible or desirable. There is a need to systematically identify the origins of natural, data, and model uncertainty in photochemical modeling applications and to develop appropriate methods for quantifying each type of uncertainty, and reducing, if feasible, data and model uncertainties. Modelers should be able to express the above quantitative estimates of uncertainty in forms that would help policy makers to implement them in the decision-making process of air quality management.

### Research Needs in the Regulatory Application of PAQSMS

The current ozone attainment demonstration guidelines reflect a rigid deterministic procedure that involves simulation of a minimum of three episodes, and demonstration of attainment in all grid cells for the domain and the days modeled. As mentioned earlier, this procedure can be made consistent with the statistical form of the NAAQS only in a qualitative way through the educated choice of episodes other than the most severe on record. The potential of developing and implementing alternative (statistical rather than deterministic) attainment demonstration approaches that are more in line with the regulatory interpretation of the standard and the ozone design value for an area should be considered in the near future. Such
approaches can be based on comparison of calculated statistical distribution attributes of ozone concentrations with corresponding statistics of observations and can utilize the fundamental concepts of order or extreme value statistics (5,41,42). For example, after a set of representative ozone episodes from a 3-year period has been identified for the domain of concern, simulations can be performed using these episodes for the base case, the future year, and the control strategy options. The maximum predicted daily maximum hourly averaged ozone concentration over the domain for each model simulation can be fitted to an appropriate empirical distribution, such as the Weibull distribution (41), and the distribution for the fourth-order statistic can be determined using extreme value theory. Then, the probability of the fourth highest maximum exceeding 0.12 ppm can be determined. Control strategy analysis would then involve simulating various emission controls and determining the corresponding distributions of daily maximum hourly ozone concentration until a specific emission reduction strategy results in an acceptable probability that the fourth highest ozone concentration does not exceed the 0.12 ppm level. This control strategy can then be considered as a potential SIP strategy. Another potential approach for assessing the likelihood of a strategy resulting in ozone attainment could be based on hypothesis testing (5,43). The t-test may provide a useful procedure for testing compliance with the ozone air quality standard. Use of such a scheme for assessing compliance with the ozone NAAQS requires a regulatory agency to make explicit decisions concerning the probability of success or what failure rate is acceptable and the probability of erroneously designating a complying area to be in violation or designating a noncomplying area to be in compliance.

It should be noted that probabilistic types of analysis will generally require simulation of a large number of episodes to obtain valid statistical distributions of ozone maxima; therefore, computational constraints may limit the direct applicability of these approaches. There is the possibility that methods of statistically enhancing sets of calculated values, through bootstrapping or similar techniques, which have been applied in conjunction with inert dispersion modeling (42), can be modified for application to photochemical modeling. However, the nonlinear nature of photochemical systems presents various problems that have to be resolved before such methods are extended to ozone modeling.

Finally, ways of extending the present hourly standard-based approach for demonstrating ozone attainment should be considered in the near future as part of a post-1994 SIP reevaluation of the objectives of ozone control. Indeed, the present form of the standard may be altered or extended in the future, e.g., to incorporate considerations relevant to longer duration exposures at lower ozone concentrations. Then, it is logical to propose that future strategy development should evolve to become more flexible and to also include other criteria in addition to reducing the maximum ozone value in an area. Such criteria may provide more robust metrics for the selection of control strategies based on the reduction of VOC and NOX emissions. Examples related to this discussion are presented in Figures 7, 8, and 9. These figures contain sample results of an ongoing diagnostic analysis for the Philadelphia—New Jersey modeling domain (1) that have been obtained with the 1988 interim U.S. EPA emission inventory and various simplifying assumptions for the meteorological inputs. Although these calculations should be interpreted strictly as components of a system sensitivity analysis, and not as indications of preferred control directions, they depict some interesting options in the context of regulatory application of ozone models. For example, calculations for the domain under consideration show that NOX reductions, starting from the base interim inventory, can potentially be counterproductive with respect to reducing domain-wide ozone maxima (Figure 7A). However, if other metrics after a certain reduction level that incorporate the impact on potential total human exposure are used, a somewhat different picture emerge. Indeed, NOX controls seem to be consistently effective (Figures 7B, C, 8 and 9) in reducing episode pervasiveness (expressed as the 24-hr sum total of the number of cells in the domain that were predicted) and episode severity (expressed as the 24-hr sum total of ozone concentrations in all cells in the domain that were predicted to be in noncompliance with the ozone standard each hour). According to these calculations, although across the board NOX controls appear to be less effective than comparable VOC controls in reducing ozone maxima for the domain of concern, they still appear useful in reducing the spatial extent of ozone episodes, and potentially, associated human exposures.

Extensive additional research is needed on these issues to provide the basis for a

Figure 7. Comparison of the sensitivity of different metrics of ozone episode severity for July 7, 1988, calculated using the ROM2.2/UAM-IV modeling system for the Philadelphia—New Jersey domain, with respect to changes in VOC and NOX emission levels: (A) % decrease of the urban domain ozone maximum for various VOC and NOX decreases relative to the base case; (B) 24-hr total of the number of cells in the domain that were predicted to be in non-compliance with the ozone standard each hr; (C) episode severity of 24-hr total of ozone concentrations in all cells in the domain that were predicted to be in noncompliance with the ozone standard each hr. These interim calculations represent part of a diagnostic analysis that is performed with the 1988 interim U.S. EPA emission inventory and various simplifying assumptions for the meteorological inputs.
Figure 8. "Tile-map" comparison of the sensitivity of the distribution of daily maximum, hourly-averaged, ozone concentrations (ppm), for July 7, 1988, calculated using the ROM2.2/UAM-IV modeling system, with respect to changes in VOC and NO\textsubscript{x} emission levels: (A) base case, (B) VOC and NO\textsubscript{x} reduced by 50\% relative to the base case, (C) VOC reduced by 75\% and NO\textsubscript{x} reduced by 25\%, (D) VOC reduced by 25\% and NO\textsubscript{x} reduced by 75\%. These "interim" calculations represent part of a diagnostic analysis that is performed with the 1988 interim EPA emission inventory and various simplifying assumptions for the meteorological inputs.
Figure 9. "Tile-map" comparison of the sensitivity of the distribution of hours of noncompliance (> 0.12 ppm) with the ozone standard, for July 7, 1988, calculated using the ROM2.2/UAM-IV modeling system, with respect to changes in VOC and NOx emission levels: (A) base case, (B) VOC and NOx reduced by 50% relative to the base case, (C) VOC reduced by 75% and NOx reduced by 25%, (D) VOC reduced by 25% and NOx reduced by 75%. These "interim" calculations represent part of a diagnostic analysis that is performed with the 1988 interim EPA emission inventory and various simplifying assumptions for the meteorological inputs.
flexible and rational regulatory framework that will identify feasible strategies for minimizing the impact of tropospheric ozone on human health.

Appendix

Operational Performance Evaluation Measures for PAQSMs (10)

Paired Peak Estimation Accuracy. The paired estimation accuracy examines the discrepancy between the magnitude of the peak one-hour average concentration four-cell weighted average determined by simple bilinear interpolation among the four grid cells nearest the monitoring location.

Temporally Paired Peak Estimation Accuracy. The temporally paired peak estimation accuracy examines the model’s ability to reproduce the highest observed concentration in the subregion surrounding the monitoring station at the same time of occurrence of the measured maximum. Relaxation of the spatial pairing requirement could be allowed up to a maximum subregional distance of 25 km.

Spatially Paired Peak Estimation Accuracy. The spatially paired peak estimation accuracy describes the discrepancy between the magnitude of the peak one-hour average concentration measurement at a monitoring station and the highest estimated concentration at the same monitor, within 3 hr of the peak. When interpreted along with other measures, it provides insight into the reasonableness of the simulated transport processes leading to the maximum concentration.

Unpaired Peak Estimation Accuracy. The unpaired peak estimation accuracy describes the difference between the magnitude of the peak one-hour average observed concentration and the highest value estimated anywhere in the modeling of region. This is the least stringent of the peak estimation accuracy measures.

Average Station Peak Estimation Accuracy. The average station peak estimation accuracy is the mean of the spatially paired peak estimation accuracies averaged over all monitoring station locations. It is calculated by first determining the spatially paired peak estimation accuracy at each monitoring station and then averaging all these values. The temporal offset between estimated and observed maximum at any monitoring station should not exceed 3 hr. The average station peak estimation accuracy describes how well the maximum concentrations throughout the monitoring network are reproduced.

Mean Bias. The mean bias (mean bias error) is calculated both as a residual quantity and one that is normalized by the observed concentrations. The bias is determined from the average signed deviation of the concentration residuals. It indicates the degree to which observed one-hour concentrations are overestimated or underestimated. Based on the ensemble of estimation-observation pairs, this measure reveals the presence of systematic deviation from observed concentrations. The nonnormalized bias is calculated to aid in developing a robust data base on photochemical model performance evaluation. The mean normalized bias, generally of greater interest, is useful in identifying systematic errors in the model’s temporal or spatial response. Since the bias reveals the tendency for systematic overestimation or underestimation, it should be zero in the ideal case. Caution must be exercised in the interpretation of bias because it is possible for large, compensating subregional biases to produce a mean zero estimate.

Variance. The variance of the distribution of residuals describes the dispersion of the residual distribution about the mean. As the second moment of the concentration residual distribution, the variance is a measure of the average spread of the residuals, independent of any systematic bias in the estimates. The variance provides no direct information about subregional errors or about large discrepancies occurring within portions of the diurnal cycle.

Gross Error. The gross error, reported as both normalized and nonnormalized measures, describes the average absolute signed deviation of the concentration residuals. It indicates the average (signed) discrepancy between hourly estimates and observations and is one of the most useful measures for comparing different model simulations. The normalized gross error is a robust measure of overall model performance, representing the average error in estimation.

Graphical Performance Procedures

Accuracy Plot. Two accuracy plots are recommended; one depicts relationships between the five numerical peak estimation measures, and the other plot provides a comprehensive summary of the peak estimation accuracy at all monitoring stations.

Time Series Plots. Probably the most useful graph for depicting photochemical model results is the time-series plot. Developed for each monitoring station for which observed concentrations are available, this plot presents the hourly estimates and observations throughout the simulation period. The absolute value of the concentration residual value is also presented on the same plot. One may determine the model’s ability to reproduce the peak estimation, the presence or absence of significant bias and errors within the diurnal cycle, and whether the timing of the estimated concentration maximum agrees with the observations. By including the residual plot on the same graph, estimation biases are more apparent.

Spatial Time Series Plots. Spatial time series plots provide information about the degree to which model discrepancies result from the procedure for selecting the estimated values. Time series plots are constructed for each monitoring station by plotting the hourly observations together with three sets of model estimates: the four-cell weighted average based on bilinear interpolation; the estimate in the grid cell containing the monitor; and the estimate closest in magnitude to the observed value, where the estimate at a given hour is drawn from one of the four nearest grid cells.

The spatial time series plot provides useful diagnostic information about the steepness of the concentration gradients in the simulated fields. Spatial time series plots are one method of revealing the commensurability between volume-averaged model estimates and point measurements.

Ground Level Isopleths. Ground level isopleths display the spatial distribution of estimated concentration fields for any selected hour. Developed by computer-contouring the hourly gridded model estimates these isopleths supply direct information about the magnitude and location of pollutant concentrations and help to identify situations were subregional biases may be attributed to spatial misalignment of the estimated and observed concentration fields.

Scatter Plots of Estimates and Observations. Scatter plots depict the extent of bias and error in the ensemble of hourly estimation-observation pairs. Bias is indicated by the preponderance of data points falling above or below the perfect correlation line. The dispersion of points is a measure of error in the simulation. Scatter plots are helpful in identifying potential outlier estimation-observation pairs. These plots provide little diagnostic information about subregional performance problems, temporal or spatial misalignments, or other inadequacies in the simulation.

Scatter Plot of Residuals and Observations. The residual scatter plots
reveal the distribution of hourly model discrepancies (positive and negative) as a function of concentration level. The plot does not reveal the existence or causes of subregional or timing performance problems. The smaller the scatter about the ordinate, the smaller the modeling error. Absence of bias is indicated by no systematic tendency for the data points to fall above or below the ordinate.

**Bias Stratified by Concentration.** The bias-concentration plot depicts the degree of systematic bias in hourly averaged model estimates (paired in time and space) as a function of observed concentration level. The bias-concentration plot reveals the existence of underestimation or overestimation within any concentration interval.

**Gross Error Stratified by Concentration.** The gross error-concentration plot depicts the degree of error in model estimation (paired in time and space) as a function of observed concentration level. The gross error-concentration plot, revealing the variation in model error at various intervals throughout the concentration range, must be interpreted carefully because the residual error is normalized by the observed concentration.

**Bias Stratified by Time.** The bias-time plot identifies specific time periods within the photochemical simulation when systematic tendencies toward underestimation or overestimation occur. The bias-time plot is constructed in a manner similar to the bias-concentration plot, except that the simulation period is discretized into a number of time intervals, usually 1 to 2 hrs in duration.

**Gross Error Stratified by Time.** The gross error-time plot identifies specific time periods when errors in the model estimates may be a problem. The gross error-time plot is constructed in a similar manner as the error-concentration plot.

### Investigative Simulations

Six investigative simulations are suggested as the minimum set of tests to accompany the above numerical and graphical procedures:

**Zero Emissions.** The purpose of the zero emissions investigative simulation is to ensure that the base case simulation results are influenced appropriately by the emissions inputs. Eliminating all emissions should lead to significantly reduced reactive species concentrations on the second and subsequent simulation days. The zero emissions simulation is performed by exercising the base case run with all emissions values reduced to zero. All other model input files remain unchanged from the base case.

**Zero Initial Conditions.** The zero initial conditions simulation quantifies how much of the second- (or third-) day estimates are the result of the initial conditions used to start the simulation. This simulation is performed by setting all initial concentration fields in the model to zero. If the initial field is completely washed out of the model domain by the second- (or third-) day simulation results will indicate essentially no differences between the investigative and base case runs on the following day(s).

**Zero Boundary Conditions.** The purpose of the zero boundary condition simulation is to quantify the influence of boundary conditions on second (or third) day concentrations, particularly in regions where the base-case estimates are highest. This simulation helps identify situations where the base-case results are driven by the boundary conditions. The zero boundary conditions simulation is performed by setting all inflow and outflow boundary values, including the region top, to zero. Unless there is reason to suspect that a portion of the peak concentration measurements within the region derive from transport from outside the modeling region, the model results should reveal little impact of the boundary conditions in the interior of the computational domain.

**Zero Surface Deposition.** The zero deposition simulation quantifies the influence of dry surface deposition on primary and secondary species concentration. The zero deposition investigative run is exercised by setting deposition velocities for all species to zero and rerunning the base-case simulation. Deposition tests have not been reported in previous model evaluation studies so the analyst has little historical information at present to serve as a guide in interpreting the results of this investigation.

**Increased Mixing Heights.** The objective of the mixing height investigative simulation is to reveal the degree to which ozone concentrations are influenced by the height of the mixed layer. At a minimum, one run is suggested in which the hourly mixing height values are uniformly increased by 50% above the base-case values. This simulation should provide a bound on the change in ozone estimates resulting from uncertainties in this input. One might choose, instead, to reduce the hourly mixing heights by 50%. The resultant increase in ozone concentrations under this scenario will typically be comparable in magnitude but of opposite signs as those for the mixing height increase case.

**Reduced Wind Speeds.** This investigative simulation entails a 50% reduction in the magnitude of the winds input to the photochemical model, thus providing an initial characterization of the ozone model’s sensitivity to ventilation. This simulation is performed by decreasing all grid-point wind components by 50% and rerunning the photochemical model. The magnitude of the hourly ozone concentrations, including the peak value, should increase relative to the base case although the percentage increase should be less than proportional with wind speed reduction.

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