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Environmental Energy Technologies Division

August 2003

This research was supported by the Assistant Secretary for Fossil Energy, Office of Natural Gas and Petroleum Technology, through the National Petroleum Technology Office under U.S. Department of Energy Contract No. DE-AC03-76SF0009

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EVALUATING UNCERTAINTIES IN REGIONAL PHOTOCHEMICAL AIR QUALITY MODELING*

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Key Words ozone, emissions controls, simulation models, atmospheric transport

■ **Abstract** This review evaluates analyses that are or may be performed to estimate uncertainties associated with air quality modeling used in regulatory planning to meet National Ambient Air Quality Standards for ozone. The sources of uncertainties in photochemical air quality simulation models (PAQSMs) are described. Regulatory requirements for evaluating PAQSM performance and uncertainty concerns not addressed through standard performance evaluations are discussed. Available techniques for evaluating uncertainties are presented. Experiences with analyses conducted most commonly are reviewed, as are those that might be used in a cohesive model uncertainty evaluation. The review concludes with a call for renewed emphasis on applying current techniques complemented by heretofore sparsely used diagnostic, corroborative, and alternative approaches and enhanced observational databases.

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I. INTRODUCTION

Tropospheric ozone forms on hot, sunny days via photochemistry that involves nitrogen oxides and hydrocarbons emitted from both human and natural sources. The complexity of ozone formation compels air quality managers to use a photochemical air quality simulation model (PAQSM) system to plan how to bring nonattainment areas into compliance with health-based National Ambient Air Quality Standards (NAAQS) of the Federal Clean Air Act (FCAA). Progress in attaining ozone standards has been slow. According to monitoring data gathered in 2000, approximately 52 million people lived in 30 metropolitan statistical areas where the highest second daily maximum concentration violated the ozone NAAQS threshold of 0.12 ppm averaged over one hour (1). Nonattainment areas include many major cities and rural/suburban settings, such as Houston, Los Angeles, and the San Joaquin Valley, in California. The new federal ozone NAAQS promulgated by the U.S. Environmental Protection Agency (EPA) in 1997 is another challenge. The new standard threshold concentration of 0.08 ppm averaged over eight hours was exceeded by the highest fourth daily maximum measured in over 100 areas encompassing a population of 119 million people (1).

The “discouraging and perplexing” (2) inability to reduce peak urban ozone concentrations inspired three federal studies (2–4) and a privately sponsored review (5). All reports noted significant modeling inaccuracies and called for their evaluation and reduction.

Purpose and Organization of the Review

This review describes analyses that are performed or may be developed to evaluate uncertainties in modeling results used to support emissions control plans to meet ozone NAAQS.^{1, 2} A description of PAQSMs and the sources of uncertainties in their results is provided in Section II. Section III defines uncertainty and sensitivity analyses, summarizes regulatory requirements for model performance evaluation, and introduces the techniques available for evaluating uncertainties in modeling.

Experiences with uncertainty analyses conducted most commonly, as well as those that might be used in a cohesive modeling uncertainty evaluation, are reviewed in Sections IV through VI; these include uncertainty evaluations using sensitivity (Section IV), diagnostic (Section V), and corroborative/alternative modeling and subjective judgment (Section VI). The review closes with suggestions for improving uncertainty analysis estimation capabilities.

Importance and Uses of Model Uncertainty Information

Uncertainty information is needed by planners who must decide what emissions controls to implement in pursuit of air quality standards. The fundamental question underlying a plan to meet ozone standards is: How much must current and anticipated future nitrogen oxides (NO_x) and volatile organic compound (VOC) emissions be reduced to meet the ozone NAAQS by a specified deadline? Models are used explicitly to generate information to meet the needs of the planner. Most important is the ability to simulate the interactions of complex chemical, meteorological, and pollutant emissions processes and to estimate air quality in the future. If used wisely, simulation is an “indispensable tool for predicting the outcomes of alternative policies” (6).

During SIP development, PAQSM are used to simulate an observed violation of the NAAQS concentration threshold for ozone. Once the base case simulation meets performance criteria specified by oversight agencies³, the PAQSM is rerun with scenarios representing emissions reductions from hypothetical controls. The modeling is said to “demonstrate attainment” when modeling results indicate that planned controls will reduce ozone concentrations to below the standard if the meteorological conditions in the simulated episode are experienced again. Similarly, changes in distant, upwind emissions sources may be simulated to evaluate the

¹An air quality improvement plan required by the FCAA when an area violates a NAAQS is a State Implementation Plan (SIP).

²Most emissions are associated with the production of energy; hence, control strategy design and air quality modeling are of consequence for national energy policy.

³EPA sets performance criteria to be met before using a modeling simulation for SIP planning [see (7)]. In California, additional criteria are set by California Air Resources Board (CARB) [see (8)].

significance of pollutant transport. As discussed below, modeling output may also be used to characterize uncertainties.

If model capabilities fall short of demands, decision makers still need information. They may rely on judgments to span information gaps and to justify decisions. This subjective aspect of planning often leads to controversy. There is also legal impetus to use models to demonstrate that decisions are neither arbitrary nor capricious. For example, in *Chevron U.S.A. versus Natural Resources Defense Council (NRDC)*, NRDC challenged the EPA approach for applying emissions controls to meet the requirements of the Clean Air Act Amendments of 1977. The U.S. Supreme Court supported EPA, stating, "the EPA should have broad discretion in implementing the policies of the 1977 Amendments [of the FCAA]" (467 U.S. 837). The opinion further states, "If Congress has explicitly left a gap for the agency to fill, there is an express delegation of authority to the agency to elucidate a specific provision of the statute by regulation. Such legislative regulations are given controlling weight unless they are arbitrary, capricious, or manifestly contrary to the statute." Planners rarely have other tools or information available to provide the bases for decisions. Model uncertainty information can reduce the need for judgment and make judgments explicit for the purposes of public debate.

Depending on the decision criteria,⁴ decisions may be facilitated by more complete model uncertainty information. Making decisions under uncertainty, planners should consider the likelihood that their plans will yield air quality goals once implemented. They need to assess risk, which is the chance of suffering harm or loss. The question that comes to mind is: What is the likelihood that ozone NAAQS will actually be met when the model indicates that planned emissions reductions will yield attainment?

The answer to this question is a probabilistic statement. Using modeling output as well as information about output uncertainty facilitates risk assessment. In addition to risk assessment and management, there are at least six uses for model uncertainty information:

1. Satisfy the regulatory requirement to demonstrate acceptable model performance.
2. Enable planners to estimate the probability of not meeting goals even though model projections indicate the goals will be met.

⁴Examples of decision criteria given by Morgan & Henrion (9), include:

1. *cost-benefit*, deterministically comparing the costs and benefits of alternatives or doing so probabilistically by incorporating uncertainty and then comparing expectations of costs and benefits;
2. *cost-effectiveness*, choosing least cost routes to goals that are not necessarily based on economic considerations; and
3. *zero or bounded risk*, decisions reducing or altogether prohibiting undesirable outcomes without consideration of costs or benefits.

3. Identify situations in which model uncertainties are greater than the needed air quality improvement. For example, when interpreting modeling results, it is important to know if changes in modeled ozone concentration due to changes in emissions are of the same magnitude, or less, than accompanying uncertainties.
4. Suggest alternative control plans that may produce comparable air quality improvement within the range of uncertainty of the modeling results.
5. Inform general planning and resource allocation. For example, guide the planning of large field studies by identifying what data to gather, as well as where and when to gather them.
6. Set research priorities to improve the characterization of complex atmospheric processes by using both uncertainty and sensitivity information to identify key PAQSM components that need improvement.

II. PAQSM AND THEIR UNCERTAINTIES

PAQSM Defined

A photochemical air quality simulation model is an attempt to "... describe a dynamic, physical phenomenon by mathematical relationships which, when combined with accurate input data, imitate the real system" (11). A PAQSM is a mathematical representation of physical and chemical processes occurring in the atmosphere and at the atmosphere/land interface; the model includes emissions, diffusive and advective transport, chemical transformation, and deposition. A PAQSM integrates our knowledge of the spatial and temporal evolution of gaseous and particulate constituents in the atmosphere. In addition to emissions and atmospheric processing, it represents the physical system comprised of topography (e.g., mountains), surface characteristics (e.g., land use and land cover), and meteorology (e.g., winds, temperatures, and clouds). The PAQSM domain may range from an urban airshed to a regional to a continental-scale area.

MODELS OF A PAQSM The key components of PAQSM are shown in Figure 1. They are meteorological, emissions, and air quality models. (The entire photochemical air quality simulation modeling system is referred to as a PAQSM and the embedded components within the PAQSM as models.)

The meteorological model uses pertinent information to generate meteorological "fields"—wind speed and direction, temperatures, and humidity—that are inputs to the emissions and air quality models. The emissions model calculates emissions from natural and anthropogenic sources. The air quality model contains descriptions of physical and chemical transformations, transport, and numerical solution algorithms.

Many tiers and types of models are embedded in the components of the PAQSM depicted in Figure 1. For example, the emissions model component will include

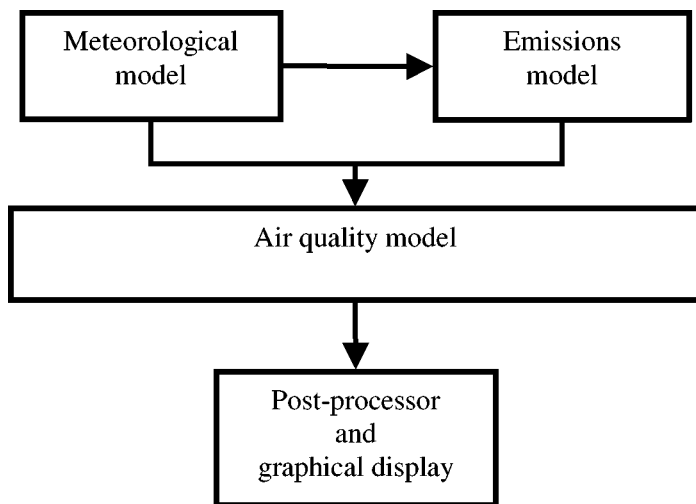


Figure 1 Component models of a photochemical air quality modeling system.

formulas for estimating emissions from stationary point, area, on- and off-road mobile, and biogenic emissions. In addition, a geographic information system may be used to organize and manipulate spatially resolved data, and post-processing systems may summarize and display results graphically.

Mathematical description of the dynamics of gases and aerosols in the atmosphere is achieved using conservation equations for mass, momentum, and energy. Pollutant transport and transformation is tracked temporally and spatially using the advection-diffusion equation (ADE):

$$\frac{\partial c_i}{\partial t} = -\frac{\partial}{\partial x_j}(u_j \partial c_i) + \frac{\partial}{\partial x_j} \left(K_{jj} \frac{\partial c_i}{\partial x_j} \right) + R_i + S_i + L_i \quad 1.$$

= advection + turbulent diffusion + reactions + sources + removal.

$j = x, y, z$ dimensions.

$i = 1, \dots, n$ pollutant species.

The ADE describes how the time rate of change of concentration, c_i , of the i^{th} pollutant equals the net changes due to five processes:

- advection by the mean wind components, u_j ,
- turbulent diffusion characterized by gradient transport using eddy diffusivity, K_j ,
- production and destruction of i through chemical reactions, R_i ,
- addition of i by emission sources, S_i , and
- removal of i at the surface or by other physical processes, L_i .

Each pollutant is described with an ADE. The result is a set of coupled, nonlinear partial differential equations that satisfy conservation relationships in a turbulent flow. Coupling occurs between pollutant species in the reaction term and is an important source of the nonlinearity of the system. The equations require numerical, not analytic, solution.

For each time step, Δt , of a simulation, pollutant concentrations in grid cells change as they are gained (lost) through inflow (outflow) and chemical formation (destruction). Transport terms include wind-induced advection and turbulent diffusion. Emissions are an inflow at the ground level or, for large point sources, into a horizontal layer aloft. Typically, emissions from tall stacks occur above the surface layer, so they are assumed to inject into a horizontal layer that is above the surface. Refer to (12) or (13) for detailed reviews of model formulation.

TYPES OF PAQSM The two types of PAQSM used today are distinguished by their frames of reference. Eulerian constructs overlay onto the modeling domain a three-dimensional (3-D) grid system of a particular resolution with a fixed frame of reference. Trajectory formulations start with a frame of reference that moves a control volume (which is often assumed to correspond to a particular air mass) over space and time using a prescribed meteorological variable, such as wind velocity.⁵

Eulerian models are the state of the science and require the least restrictive assumptions. In the United States, they are the models of choice for regulatory applications. However, they are computationally and data intensive. Oftentimes, there are insufficient data to support Eulerian model applications. Computational requirements may be a concern when considering the execution of a large number of simulations to fully explore model sensitivity or control strategy issues.

Trajectory models are less demanding computationally, require less input data, are a simpler representation of the physical system, and provide less information about the spatial character of pollutants. They use averages of observed wind speed and direction to transport a single air mass over space and time and, thus, have limited ability to represent complicated pollutant transport situations. The domain may be described as a single box, a zero-dimensional model (0-D), or as many boxes stacked vertically, a one-dimensional model (1-D), to allow for vertical mixing and pollutant concentrations that vary with altitude. The simplifications of trajectory models render them inappropriate for simulations extending beyond the period over which assumptions are valid. In particular, the integrity of the air column or boxes is violated over space and time in the presence of significant wind shear.

Many model uncertainty evaluations have been conducted using trajectory models because of their computational and input simplicity advantages, though recent efforts have used Eulerian models (14–20). It is easier to work with trajectory models, but their simplifying assumptions are violated more readily. Eulerian models

⁵Trajectory models are often referred to as Lagrangian formulations. The term is not used here because, strictly speaking, trajectory models are a simplification of Eulerian models that treat horizontal transport and turbulent diffusion as negligible.

are the dominant tools used for regional-scale air quality planning.⁶ There is a trend toward simulating larger domains (i.e., regional and subcontinental-scales) and longer time periods (i.e., an entire ozone season lasting several months). Using larger domains reduces errors associated with boundary conditions and allows for the examination of pollutant transport over regional scales. Modeling a full ozone season addresses concerns about the representativeness of simulating only one or a few multi-day ozone episodes.

PAQSM INPUTS PAQSM inputs may be categorized according to meteorology, emissions, topography, grid structure, and atmospheric concentrations specified at the outset of the simulation and at the boundary of the domain (12). The delineation of outputs and inputs is confounded by intermediate products; output from one model of a PAQSM can be input for another. Aerometric observations may be used to specify initial and boundary conditions, and to provide comparative data for model performance evaluation. The emissions and chemistry models rely on output from the meteorological model because chemical reactions and some emissions rates vary with actinic flux and air temperature.⁷ Meteorological fields, emissions estimates, and chemical kinetics and rates (i.e., chemical mechanism) are inputs for the air quality model.

PAQSM OUTPUTS Outputs are defined here as information produced by a PAQSM, notably estimated pollutant concentrations. Modelers use output, as well as intermediate products, to evaluate model performance. Policy makers are interested in predicted pollutant concentrations to determine “the emissions reductions needed to achieve the desired air-quality standards, such as the NAAQS for ozone” (2). The review by North American Research Strategy for Tropospheric Ozone (NARSTO) (4) identified four types of output:

1. Ozone concentrations estimated in space and time that result from estimated historical, current, or anticipated emissions.
2. Ozone precursors—concentrations of precursor (e.g., NO_x and hydrocarbons) or indicator species (e.g., CO) estimated in space and time. These chemical species are relevant and important, because they must be simulated accurately to conclude that the accuracy of ozone predictions is due to correct representation of relevant processes.
3. Ozone sensitivity—changes in ozone due to changes in precursor emissions or concentrations at the boundary.

⁶Point source dispersion models enjoy wide application for individual project permit applications. Because this review is concerned with regional-scale air quality planning, dispersion models are not reviewed.

⁷Although the dependency of emissions on meteorology is treated in PAQSM, feedback effects of chemical pollutant dynamics on meteorology are not. To the extent that emissions do not change dramatically, this assumption does not represent a significant source of error (12).

4. Transport—relative contributions from distant and local precursor emissions to peak ozone concentrations.

PAQSM Uncertainties

The uncertainties described in this section are found in the component models of a PAQSM, and they interact both within and across models. For organizational convenience, uncertainties pertaining to inputs, formulation, variability, and the use of results are discussed separately.

UNCERTAINTIES IN INPUTS These uncertainties are described below in terms of emissions, observational data, meteorology, chemistry, and resolution.

Emissions Estimates of emissions are among the most uncertain inputs of PAQSM (2, 4, 12). Emission accuracy determines estimation accuracy more than the choice of model or grid structure (12), although design features also matter [for example, see (21)].

Emissions from major industrial stacks are reasonably well known, with continuous monitors providing real-time emissions measurements for major facilities. This is not the case for emissions from residential, commercial, mobile, and biogenic sources. Industrial emissions from sources other than smokestacks (e.g., leaky pipes and valves) are not known accurately. Traditionally, motor vehicle and biogenic VOC emissions have been under- and overestimated, respectively [for examples, see (22) and (23)]. Russell (12) discussed how emissions estimates differ from actual values:

- Motor vehicle VOC exhaust emissions are underestimated by a factor of 2 to 4 (24–26).
- Biogenic VOC emissions are uncertain by a factor of 3 or more (27, 28).
- Other VOC sources, if studied in more detail, would be found to be very uncertain too.
- Mobile NO_x emissions are better understood than mobile VOC emissions (24, 26).
- Biogenic NO_x emissions estimates are still being developed and may be important in some areas [see (29)].

Adding to emissions' uncertainties is the need for their temporal and spatial specification. In one recent study, a fuel-based estimate of diesel truck emissions for the San Francisco Bay and San Joaquin Valley air basins found that emissions decrease 70%–80% on weekends (22). Emissions variation with meteorology is quite complicated, because changes in photolytic flux, temperature, and moisture can influence emissions (30, 31). Efforts to distinguish weekday and weekend emissions budgets are ongoing but hindered by limited knowledge of day-to-day changes in anthropogenic activity.

Emissions estimates typically have less spatial resolution than needed for a gridded modeling application (12, 26). All emissions, except some major point sources, are treated as area sources emitted at the surface because they are summed within grid cells and assumed to mix instantaneously in the cell. This may cause an important distortion in the chemistry in the case of large point sources (12). Assuming instantaneous pollutant mixing within grid cells has the effect of slowing down the chemistry at the source, where concentrations will be higher in reality, and speeding it up at distant parts of the grid, where concentrations would be lower in reality. Plume-in-grid modeling allows for simulation of subgrid, point source emissions, where the emissions plume is assigned to the appropriate horizontal layer based on consideration of plume rise and meteorology. Overlapping plumes are not treated explicitly in such plume-in-grid models.

Biogenic emissions estimates rely on knowledge of the surface area coverage of plant types, indices of leaf mass per plant type, and emissions rates per unit leaf mass per plant type. This is an active field of research because emissions rates are characterized incompletely for the myriad plant types and VOC species emitted.

Important assumptions used to estimate future emissions pertain to the rates used for population and economic growth and for land use conversion, forecasted changes in driving patterns, and the anticipated effectiveness and rates of implementation of emissions control technologies. Inevitably, these assumptions will lead to some error. None of these assumptions can account for unanticipated gradual changes, such as the rise in popularity of sport utility vehicles and light-duty trucks during the 1990s, or abrupt changes, such as a sudden increase in crude oil prices that leads power producers to switch from oil to natural gas fuel.

Observational data Observational data collected to initialize the modeling system, provide boundary conditions, and evaluate model performance have uncertainties due to limited characterization of their spatial and temporal variability. Observational data also have uncertainties caused by monitoring equipment, user error, and monitoring network design. Some pollutant species are easier to measure than others. For example, measurements of NO_x may actually capture NO_y , which includes NO_x plus products of NO_x oxidation.⁸ Equipment may malfunction or may not be properly calibrated. Concentration estimated using canister samples may have errors due to a flawed analytical technique.

Monitor location can affect measurement bias. Routine monitoring stations are often located near sensitive receptors (e.g., population centers) or to emphasize some emissions sources over others. For example, stations may be sited near roadways to observe carbon monoxide hot spots. Measurements of ozone taken near roadways are underestimates of larger spatial scale concentrations due to local scavenging by vehicular nitrogen oxide emissions. Routine monitoring rarely

⁸ NO_y may consist of HNO_3 , HONO , N_2O_5 , NO_3 , peroxyacyl nitrates (PAN), organic nitrates, particulate nitrates, and any other reactive nitrogen compounds present (13).

characterizes ambient conditions aloft with sufficient spatial resolution, even though such observations are needed to initialize multilayer models.

Another concern is incommensurability between the spatial scales of estimated and observed pollutant concentrations. Measurements taken at one or several points within a grid cell typically represent volumes on the order of tens of cubic meters. Modeled concentrations are grid-cell averages for volumes ranging up to several hundred cubic kilometers (4). Surface layer grid cells range from 4 to 40 km in the horizontal and 50 to 200 m in height, which equals a volume range from less than 1 to 320 km³.

Meteorology The meteorological model relies on observations typically lacking in the spatial and temporal detail needed to initialize meteorological fields. Interpolation using sparse observational data can lead to errors in calculated meteorological fields. The state of the science has progressed from objective interpolation to prognostic methods based on solution of fluid dynamics equations. Errors associated with initial and boundary conditions and with numerical solution methods can amplify temporally (32). The application of four-dimensional data assimilation dampens the temporal growth in errors by causing model results to conform to observations at regular intervals (33). Specifically, the three components of wind velocity calculated as a function of time are nudged toward measured values. Doing so, however, reduces the amount of observational data remaining for performance evaluation.

An important characteristic of meteorology is solar radiation, which influences temperature, photochemical reactions, and vertical mixing (12, 34, 35). Radiative transfer depends on incoming solar radiation, scattering and absorption by gases, aerosols and ground-level surfaces. Surface albedo influences actinic flux, which must be known to estimate chemical processing. The effects of aerosols on radiative transfer, both direct and indirect (i.e., due to clouds), are major sources of uncertainty. Actinic flux estimation requires description of aerosols spatially and temporally; only recently are observations available to do so accurately. There remain large uncertainties in aerosol concentrations, composition, and optical properties. Representations of key cloud chemistry and physics are also limited. With the exception of NO₂, atmospheric gases that affect radiative transfer are better characterized, and there are fewer uncertainties associated with their contributions.

Chemistry Atmospheric chemistry is known incompletely because it involves hundreds of pollutant species and thousands of reactions. Reaction rates and pathways are understood adequately for less than one quarter of the chemical species observed in the atmosphere (4). Even if known completely, atmospheric chemistry cannot be represented in its entirety because it would impose excessive computational demands. Fortunately, only a subset of essential reactions need be represented. Several approaches are used to simplify the chemistry, among these are the steady-state approximation for radical species and the use of structural and

functional lumping for organic species (36, 37). Although they give similar results for ozone, they do not do so for other pollutant species (38). Uncertainties associated with atmospheric organic chemistry are more significant when ozone formation is limited by the availability of organic compounds; that is when the ratio of VOC to NO_x is small. Chemical kinetic parameters are determined experimentally, so the values are subject to experimental errors.

Resolution Representing the range of scales relevant to the physical system places great demands on a PAQSM (39). Models must span orders of magnitude in time and space, even though outputs are usually sought for time periods covering hours or days.⁹ A compromise must be met between the inherent resolution of the processes of interest and scales imposed to manage the limitations of available information and computational intensity.

The spatial and temporal resolution of the inputs from the meteorological and emissions models are determined by modeling system specifications, such as grid structure (i.e., horizontal and vertical resolution). Emissions estimates and meteorological field inputs must be resolved to these scales. Furthermore, emissions descriptions must be compatible with the speciation requirements of the chemical mechanism.

Chemical and meteorological measurements are often not available at desired temporal or spatial resolution. No matter what grid size is selected for use, processes occurring at subgrid spatial scales are represented by average values used for grid cells. The significance of this limitation is still being explored (4).

There is a trade-off between grid structure resolution and computational intensity. Finer grids, both horizontally and vertically, reduce errors associated with numerical solution techniques, better represent point-oriented observations, and facilitate the approximation of physical processes, such as wind shear and vertical mixing. Data availability becomes limiting because finer resolution is not helpful when inputs, such as observations and estimates of emissions and meteorological variables, are not similarly resolved. New approaches, e.g., nested-grid refinements (40–42), might address this trade-off by providing finer spatial resolution for emissions hot spots.

UNCERTAINTIES IN MODEL FORMULATION Uncertainties associated with model formulation may be due to erroneous or incomplete representations, incommensurability, numerical solution techniques, and choice of modeling domain and grid structure. Simplified representations are necessary when knowledge is incomplete, or when more thorough or precise specification would increase computational intensity excessively. If more than one algorithm is available and appropriate, choosing one inevitably means accepting some uncertainties over others.

⁹The averaging times for ozone NAAQS concentration thresholds are one and eight hours.

Turbulence Uncertainties arise from the deterministic representation of turbulent diffusion transport using the gradient transport hypothesis in conjunction with the diffusivity coefficient, K_j . The approach limits the model applicability in the lower limits of the spatial and temporal scales (12). The validity of the ADE is predicated on two assumptions. Atmospheric turbulence is assumed stationary for the averaging time period of interest (~ 30 to 60 minutes for most applications). Also, the characteristic temporal and spatial scales in the gradients of the turbulent velocity correlations are assumed large compared with the time resolution and average distance that a fluid particle travels in that time period. These assumptions break down at small spatial and temporal scales.

Removal processes Uncertainties in estimating pollutant removal are associated with the treatment of pollutant transport near surfaces and the net flux of pollutants from various types of vegetation and soils. Deposition is the pathway by which pollutants are removed from the atmosphere via the physical transport to the surface and the physical/chemical interactions that occur there. The nature of these interactions for various species and surface types is a source of uncertainty. In wet conditions, deposition involves washout of pollutants with precipitation. Dry deposition involves no atmospheric hydrometeors (i.e., cloud and fog droplets, rain, or snow).

Available measurements of deposition are limited (43). Studies of the processes that control dry deposition require direct measurements of the air-to-surface exchange. Micrometeorological approaches used to characterize deposition, e.g., eddy correlation and gradients, are not well developed for nonuniform landscapes, such as hilly terrain, or for reactive pollutants, such as NO_2 . The ability to parameterize the processes affecting dry deposition and reemission is limited by the complexity and variability of the chemical, physical, and biological characteristics of the surfaces and the diversity of pollutants and surface types.

Wet deposition and other aqueous phase physical and chemical interactions are among the most complex atmospheric processes to model. Indicative of the challenge are the range of scales (i.e., 10^{-6} m to 10^6 m) at which relevant processes occur, the multiple phases (i.e., gas, liquid, and aerosol), states of aqueous phases (e.g., cloud droplets, fog, rain, snow, and ice), and different processes occurring within and below clouds (13). For simulations involving short-term and urban-scale ozone episodes in areas of low humidity (e.g., western United States), wet deposition is not important because high ozone typically occurs on dry days. However, in humid areas (e.g., midwestern and eastern United States) and for regional-scale, seasonal-length modeling, precipitation scavenging and cloud dynamics become significant. These processes are treated using a washout parameter determined empirically or by calculating rates of pollutant diffusion into water droplets. Simulation is hindered by deficient or inaccurate knowledge of the size distributions of water droplets and ice crystals, as well as incomplete understanding of cloud dynamics. Consequently, wet deposition is one of the more uncertain outputs of meteorological models (44).

Another removal process is the entrainment of pollutants aloft (i.e., above the mixing layer). As modeled ozone episodes may last many days, overnight storage and subsequent reintroduction of aloft pollutants require description of the rate of vertical mixing. Doing so introduces uncertainty because knowledge of turbulent flow, vertical exchange, and pollutant concentrations at the top of the modeling domain is usually limited.

Aerosols Historically, models sometimes treated the transport of aerosols but never their physical and chemical processing. Gas and liquid phase chemistry requires specification, as does the chemistry involving pollutant reactions with aerosol and water droplet surfaces. This is especially so when simulating regional spatial scales and entire ozone seasons. Cloud droplets act as small reactors, influence pollutant mixing, compete with gas phase chemistry, and affect rates of wet deposition. Knowledge of heterogeneous (multiphase) reactions is severely deficient. Treating cloud processes is computationally intensive and requires input data that are rarely available. Complete treatment requires characterization of the size distributions of aerosols as well as their other chemical and physical properties. Consequently, there is considerable uncertainty associated with the treatment of cloud dynamics by deterministic meteorological models. Typifying how uncertainties are interdependent, those associated with cloud predictions exacerbate uncertainties in biogenic emission estimates that are sensitive to photolytic flux.

Numerical solution Solution techniques pertain to the numerical methods used to solve the set of coupled differential equations that cannot be solved analytically. Each of these conserves mass approximately (12). Solution-related errors tend to resemble artificial dispersion, thereby spreading would-be concentration peaks spatially. Nonetheless, solution techniques are believed to contribute a small amount of error to model predictions relative to errors associated with emissions estimates, representation of meteorology, and values used for boundary conditions (45).

VARIABILITY Variability refers to stochastic atmospheric and anthropogenic processes. It contributes to uncertainties discussed previously, notably those associated with emissions estimates and representations of chemistry and meteorology. Here, its contribution to uncertainty is discussed in two respects: the implications of using means to represent values that vary and the inability to treat inherent variability.

The deterministic treatment of stochastic processes using nominal mean values is a source of uncertainty. For example, real motor vehicle driving activity and associated emissions vary over time, e.g., daily, hourly, monthly, annually. Attempts to estimate vehicular emissions introduces uncertainty associated with the choice of representation. Although it is desirable to generate separate estimates for weekdays and weekends, available information may not be adequate to do so.

Furthermore, when mean values are used, simulation of extreme realizations is prohibited (4).

Although it may be possible to represent stochastic processes using probabilistic methods, doing so does not eliminate the uncertainty inherent to variability. The estimate of vehicular emissions does not associate weather conditions and driving, so feedback effects are not represented. Extreme events are not represented either, such as changes in emissions from congestion caused by a traffic accident.

With few exceptions [for example, see (35)], neither the modeling systems nor the air quality planning efforts using modeling tools have incorporated representations of variability. At the modeling system level, emissions and meteorology are not characterized probabilistically. At the planning level, the few simulated ozone episodes may not represent the myriad conditions capable of causing violations of ozone air quality standards. With increases in computing capabilities and expanded observational databases, efforts are now under way to model full ozone seasons [see (46, 47)]; these presumably capture numerous ozone episodes to address concerns about meteorological and emissions variability. It may be necessary to model several ozone seasons to assess fully the range of variability and to evaluate air quality on days when ozone violates the 8-hour concentration threshold in addition to the 1-hour threshold. Doing so, however, may involve trade-offs. Although variability may be better represented, each modeled episode may be less accurate because it will receive less detailed attention and be based on routine rather than intensive observations.

UNCERTAINTIES IN USE OF MODELING RESULTS Although not strictly within the scope of this review, a fourth uncertainty relates to the use of PAQSM results. Decision makers must decide what to do with model output, which includes weighing it against other information. PAQSM output may be not compatible with the needs of decision makers. Consequently, there is uncertainty about how PAQSM output will be incorporated into decisions.

Another aspect of uncertainty arising from the use of model outputs is the characterization and incorporation of uncertainty. To date, formal model evaluation efforts have been inadequate (2, 4, 12, 39). Air quality planning oversight agencies provide limited guidance for treating uncertainty. Consequently, there is uncertainty about the nature of PAQSM output uncertainties, as well as how policy makers manage the limited knowledge they do have about uncertainties.

Finally, the possibility of uncertainties that are not yet known must be acknowledged. Unknown unknowns have frustrated past modeling and planning, and may continue to do so, as exemplified by the continual discovery of new sources of emissions.

Experiences with PAQSM Uncertainties

Now that models and their sources of uncertainty are described, experiences with them are discussed to indicate the potential benefits of rigorous uncertainty

assessment. In one example, Hanna (20) queried experts to estimate the uncertainties of 128 key input variables of a modeling application. The experts were instructed to describe 95% confidence intervals. The modeling system in question was used by the Ozone Transport Assessment Group (OTAG) to evaluate emissions reductions needed to bring the northeastern United States into attainment with the ozone NAAQS. The effort was particularly concerned with long-range transport of emissions from midwestern states (48). Findings of Hanna (20) are listed in Table 1.

Although indicative of the range of input uncertainties, the study of OTAG modeling by Hanna (20) is not representative of modeling carried out elsewhere. [Hanna (20) used the estimates of input uncertainties to generate an estimate of output uncertainty. The results and limitations of that effort are discussed below.] Roth et al. (49) assess eleven urban-scale and four regional-scale modeling

TABLE 1 Experts' estimates of model input uncertainties (20, 86)

PAQSM input category	Input variable	Uncertainty range	Standard deviation (log-normal distribution unless noted)
Initial conditions	O ₃ concentration	Factor of 3	0.549
	NO _x or VOC concentration	Factor of 5	0.805
Boundary conditions	O ₃ concentration aloft or at side	Factor of 1.5	0.203
	NO _x or VOC concentration aloft or at side	Factor of 3	0.0549
Meteorology	Wind speed	Factor of 1.5	0.203
	Wind direction	+/- 40 degrees	20.0 (normal)
	Air temperature	+/- 3 K	1.5 (normal)
	Relative humidity	30%	15.0 (normal)
	Daytime vertical diffusivity below 1000 meters	Factor of 1.3	0.131
	Nighttime vertical diffusivity at all other times and heights	Factor of 3	0.549
	Rainfall amount	Factor of 2	0.347
	Cloud cover	30%	0.15 (normal)
Emissions	Cloud liquid water content	Factor of 2	0.347
	Major point source NO _x or VOC	Factor of 1.5	0.203
	All other emissions estimates ^a	Factor of 2	0.347
Photolysis rates	Six reactions	Factor of 2	0.347
Carbon bond IV reactions	Reactions 1 through 83	Factors ranging from 1.17 to 2.5	0.079–0.458

^aIncludes NO_x and VOC emissions estimated for biogenic, mobile, and area sources.

applications in the United States using 20 criteria describing the soundness of the model formulation, representativeness of the modeled episode(s), adequacy of the input database and emissions estimates, and the sufficiency of performance evaluation, peer review, and documentation. Modeling limitations were found to be extensive and ubiquitous. The average difference between observed and modeled 1-hour ozone concentrations ranged from 20%–35%. Although the regional studies tended to be more satisfactory than urban-scale studies, most were found to have some or major deficiencies and omissions. Furthermore, no study performed an adequate or satisfactory estimate of modeling uncertainties.

The work by Hanna, Roth, and colleagues indicates a commitment to acknowledge and evaluate uncertainties in PAQSM in the research realm. Although the findings of Roth et al. (49) indicate less interest in uncertainty for regulatory applications, such efforts certainly benefit from progress made by researchers. In addition to improvements noted above pertaining to meteorological and plume-in-grid modeling, the NARSTO Assessment (4) highlights several modeling advancements during the 1990s, which include:

- variable grid-size nesting to permit a range of spatial resolution within the modeling domain,
- improved treatment of biogenic emissions, including isoprene estimate accuracy and incorporation into chemistry representations, and
- progress toward multi-pollutant modeling, notably the development of algorithms to represent aerosol dynamics.

Furthermore, the NARSTO Assessment acknowledges improved approaches for conducting sensitivity analyses and for estimating error distributions of model outputs. These approaches are reviewed below after the suite of methods available to evaluate uncertainties in modeling is introduced.

III. FRAMEWORK FOR UNCERTAINTY ANALYSES

Uncertainty Analyses Defined

Uncertainty analysis is defined by Morgan & Henrion (9) as “the computation of the total uncertainty induced in the output by quantified uncertainty in the inputs and model, and the attributes of the relative importance of the input uncertainties in terms of their contributions.” This approach involves sensitivity analyses that estimate output dependence on inputs, formulations, or design features (e.g., grid resolution). Sensitivity analyses are a component of the broader framework defined here as uncertainty analyses. When sensitivity studies are used to estimate total output uncertainty due to input uncertainties, it is sensitivity/uncertainty analysis.

The framework presented in this section provides the information per Morgan & Henrion (9), but it also examines performance in terms of intermediate products

(e.g., meteorological fields) and includes the use of data and models exogenous to the PAQSM. Products of uncertainty evaluation may be qualitative or quantitative.

A comprehensive model uncertainty evaluation will:

- quantify model sensitivities, notably the dependence of outputs on local changes in inputs, formulations, and design features, such as grid resolution;
- provide information to make probabilistic statements about the indications of model output, notably the likelihood that future air quality estimated by the model will be realized;
- increase confidence that the model is sufficiently valid for the decision-making need by identifying and correcting bias; and
- identify and assess the significance of compensating errors.

Before introducing the components of a comprehensive model uncertainty analysis, current regulatory requirements for performance evaluation are summarized to make a case for more extensive uncertainty assessment.

Operational Performance Evaluation

State and federal agencies establish formal processes for model validation, verification, and application. The EPA established guidelines on which PAQSM to use and how to apply and evaluate them (7, 50–54).¹⁰ In California, CARB provided similar guidance (8). EPA and CARB requirements constitute an “operational analysis” that relies on comparison of estimated and observed peak ozone, expressed in terms of bias and error metrics.¹¹ Although necessary measures, these metrics are not sufficient indicators of reliable model performance. They do not address the concern that models may appear accurate for the wrong reasons. Errors that offset each other (“compensating errors”) may indicate, incorrectly, adequate model performance. The risk of having such errors present is the development and adoption of ineffective or counterproductive emissions control strategies.

Biases or compensating errors are often hard to detect unless they are sought. The NARSTO Assessment offers two examples of compensating errors. In Kern County, California, two studies using the same model and input data produced conflicting conclusions about the relative effectiveness of pursuing NO_x or VOC control strategies, due to different assumptions about VOC concentrations aloft

¹⁰Additional guidance is provided for development and use of input data, notably emissions estimates [for examples, see (55–57)].

¹¹Here, error and bias are defined strictly. Error is the mean of the absolute values of the differences between the computed (i.e., model estimated) and observed values; bias is simply the mean of the differences. Minimum performance criteria for PAQSM used in State Implementation Plans are that peak ozone predictions have paired bias and error normalized for peak ozone of less than 15% and 35%, respectively, and a bias unpaired in space and time of less than 20% (8).

and meteorological conditions in areas without observations (4). In another example, a modeling study in New York State yielded considerable differences in ozone estimates depending on the method used to generate wind fields and mixing heights (58).

Performance criteria measured in terms of error and bias metrics appear to be based largely on experiences with model performance (4). More relevant criteria would relate to what is meant to be learned by the modeling effort. For example, the needed accuracy in estimates of observed peak ozone concentrations might be determined by the amount of ozone reduction necessary to meet the standard.

Criteria based on past performance may lead to an approved model incapable of estimating future ozone with adequate reliability. Ozone control plans based on such models may be similarly unreliable. This concern is exacerbated by lower absolute values being modeled as peak observed ozone declines and by the lower concentration threshold of the new 8-hour ozone standard.

Statistical measures tell a partial story, one that is quite helpful when there is a big problem associated with performance, but less so (from the standpoint of uncertainty estimation) when there is not. Concerning limitations of operational evaluations to estimate uncertainty, one must consider that:

- Uncertainties associated with both observed and modeled concentrations should be taken into account.
- Some measurements do not compare exactly with the quantity modeled. Per the discussion of incommensurability above, observed and modeled concentrations represent different spatial averaging characteristics. In another example that also indicates uncertainties in measurements, what is often considered observed NO_2 will likely contain some peroxyacetyl nitrate (PAN) and HNO_3 as well.
- The error and bias metrics used to summarize model performance indicate how well the model simulates the observed conditions, but they do not account for the possibility that the observed value may not be the true value of interest (e.g., peak ozone observed may not be the peak occurrence).
- Estimates of models may not be best estimates, and their expected bias should be considered. For example, ozone concentrations observed near roadways should be depressed relative to a model-estimated value due to reaction of ozone with NO . A less biased comparison would be made between observed and modeled “ozone + NO_2 .”

An unbiased, integrated estimate of uncertainty would take into account these additional factors. The NARSTO (4) and National Research Council (NRC) (2) assessments call for model uncertainty evaluations that extend beyond the criteria currently in force. Other researchers note that the practice of PAQSM performance analysis is wanting [for examples, see (12, 39, 49, 59)]. This review echoes those concerns.

Framework for Developing Uncertainty Information

Although CARB and EPA standardized the metrics for evaluating the adequacy of PAQSM performance, no comprehensive approach exists for estimating uncertainties in the results or outcomes of modeling (60, 61). Oftentimes uncertainties are not estimated. When they are, a necessarily limited approach is taken, such as the operational analyses just described. A typical uncertainty assessment will generally address one (or more) aspect(s) of uncertainty, but it will not provide a complete or encompassing estimate. The modeling community simply does not know how to do this (4).

Figure 2 introduces the several approaches now in use and shows how they might interact if applied in unison. Although not shown in the figure, a “code audit” or scientific evaluation of the process representations is another component of model evaluation (4).

Once the results of a given approach emerge, it is usually difficult to engage them with other sets of results. Methods of integration and synthesis are yet to be developed. The individual approaches represented in the diagram are:

- operational evaluation involving calculation of standard performance statistics,
- diagnostic analyses conducted typically to identify ways to improve operational performance and to assess the reasonableness of the representation of key atmospheric processes,
- sensitivity and sensitivity/uncertainty analyses,
- corroborative methods, and
- subjective judgment methods.

Assessing and estimating PAQSM uncertainties involves exercising the entire PAQSM or components of it. Modeling results may be corroborated through other means too. Diagnostic assessment focuses on individual models of the PAQSM. Corroborative methods include model-to-model comparisons, corroboration using different types of models (e.g., the EPA Mapper), data analyses, and executing the PAQSM with different scenarios of, for example, emissions estimates. Subjective judgment methods involve the solicitation and aggregation of expert opinions.

IV. SENSITIVITY ANALYSES

Overview

Model sensitivity analysis estimates model responses to changes in the model. When the effect of changing model inputs is sought, analyses are referred to as either parametric or functional sensitivity analysis; the respective distinction depends on whether the input in question is a constant or is distributed in space or time. When the effect of changing the representations of the chemical and physical

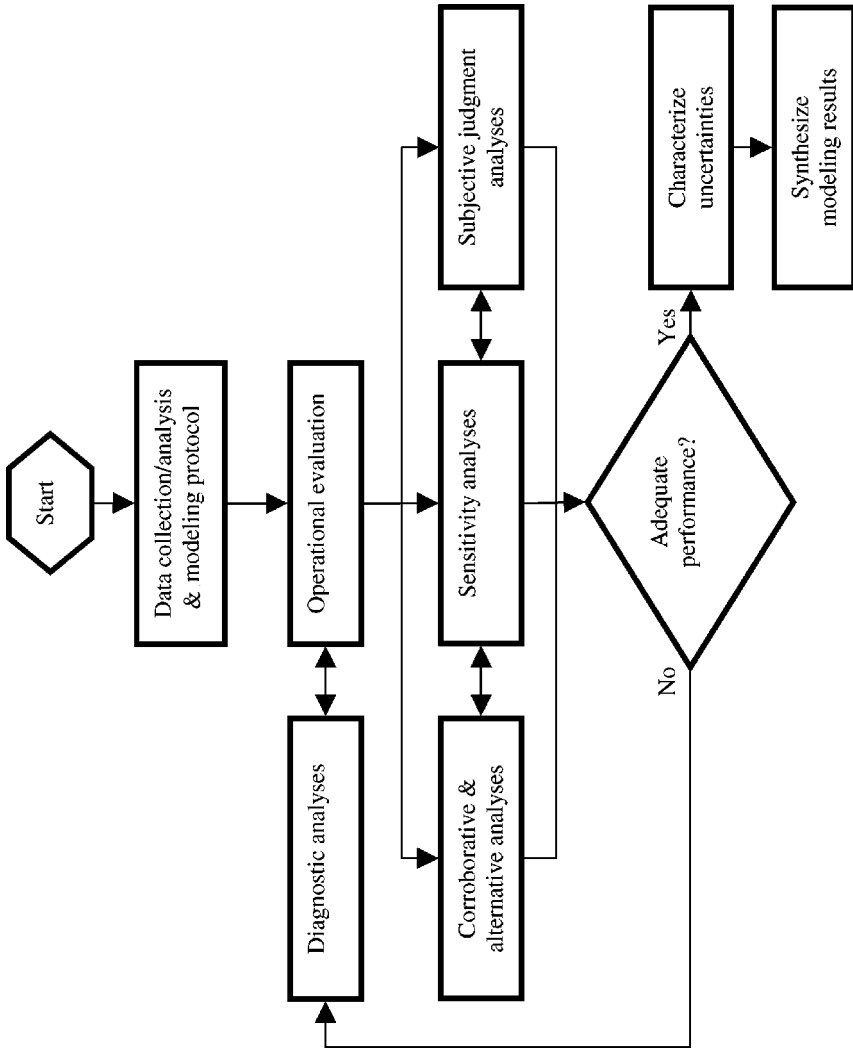


Figure 2 Comprehensive evaluation and analyses of PAQSM uncertainties.

processes is sought, analyses performed are structural sensitivity analyses. In this section, the use of sensitivity analysis for uncertainty estimation is discussed, and the constraints imposed by model formulation and by our knowledge of important input parameters are noted. So too are the various methods for computing sensitivities and uncertainties and pertinent examples of their application.

PARAMETRIC AND FUNCTIONAL SENSITIVITY ANALYSIS Parametric sensitivity analysis involves the calculation of system gradients, $\partial o_i / \partial z_u$, where o_i is the i^{th} -dependent variable (i.e., output) and z_u is the u^{th} input parameter. The system gradients describe the relationship between model responses and inputs. Referred to as sensitivity coefficients, the gradients reveal the importance of the various model parameters at a chosen point of operation. Sensitivity coefficients are quantitative measures of the significance of input parameters to outputs. Coefficient signs indicate directional response.

In air quality modeling, the dependent variables of particular interest are concentrations of pollutant species, such as ozone. Their sensitivities to input variables constitute the elementary sensitivity coefficients. Individual rate coefficients, emissions estimates, boundary conditions, and other input parameters are independent variables to be explored through sensitivity analyses. The aim is usually to identify the significance of the parameter. Often the product of an observable's sensitivity coefficient and a parameter uncertainty is taken as a measure of the parameter contribution to uncertainty in the observable. Additionally, the effect of neglected parameters (after assuming a nominal value) can also be determined by computing sensitivity coefficients for them.

When the parameters are distributed in space or time, functional derivatives, $\delta o_i / \delta z_u$, are the basic response functions associating incremental variations, δz_u , to corresponding changes in outputs, δo_i . An example of this is a chemical rate coefficient that varies with spatially and temporally varying temperature. Although functional sensitivity analysis has been used in other fields (62), this powerful technique has been applied to few air quality modeling studies.

One noteworthy example is Cho et al. (63), who used functional derivatives to explore the relationships among emissions sources, regional air quality, and acid deposition for the advection-diffusion equation. Functional derivatives are particularly useful for investigating source/receptor relationships in situations where the emission sources are distributed spatially and temporally within the modeling domain. The technique enabled Cho et al. to determine the region and magnitude of the influence of specific emission sources. The work by Cho et al. was undertaken in the late 1980s when the additional computational burden imposed by the calculation of sensitivity densities posed a more significant barrier than it would today with parallel computers. Consequently, the technique most certainly should be revisited.

Sensitivity analyses are often classified as local and global. Local sensitivity coefficients are the gradients about the nominal value of the input variables. They may be partial or functional derivatives and may include higher-order derivatives.

The key is that they are computed with respect to the nominal values of the parameters that define the problem. Oftentimes local methods are used to determine uncertainties in the dependent variable. Doing so requires variations in the input variable small enough to use a low-order Taylor series expansion or a first-order sensitivity coefficient of the dependent variable to describe the uncertainty. Too frequently modelers fail to verify that a first-order expansion is adequate.

For large variations, global methods must be used that take into account the range and statistical properties of the input variable to calculate average sensitivities, $\langle \partial o_i / \partial z_u \rangle$, over the range of the input parameter uncertainty. The simplest approach is repeatedly rerunning the model with variations of the parameter of interest. Parameter variations also may be generated systematically using factorial design and response surface approaches. Global analysis is difficult to achieve in practice because it presupposes reliable knowledge of the uncertainty in an input variable. Should one attempt a global analysis, an efficient way to conduct it would be to use local and global methods in concert. The local approach could be used to identify influential parameters to be subjects of a global analysis. Although not a formal global analysis, in the sense that average gradients associated with variations of specific parameters are not calculated, many applications of sensitivity/uncertainty analysis involve parameter variations that are larger than those associated with first- or second-order local analysis.

In addition to the various approaches to sensitivity analysis, there are myriad applications. Though several reviewers discuss applications (64, 65), Rabitz et al. (66) is particularly thoughtful in enumerating the uses for sensitivity analysis:

1. identifying influential or extraneous parameters and processes to deduce influence from the magnitude and sign of the sensitivity coefficient;
2. quantifying the extent and sources of error using a simple product of the sensitivity coefficient and an assumed error in a parameter to deduce the contribution the parameter makes to an observable's error;
3. identifying missing model components by allowing a parameter to be included in the model with zero value and computing its sensitivity coefficient, which, if large, suggests the parameter should be included in the model;
4. mapping parameter space for functional analysis whereby spatial and/or temporal regions of greatest influence are identified;
5. fitting a model to data to identify experimental conditions in which model components of interest are most sensitive to data; and
6. identifying steepest descent paths for optimization calculations to help search a multidimensional surface efficiently.

Applications 1 and 2 have been used extensively in air quality modeling. For example, sensitivity analysis is often used to determine how pollutants of interest respond to changes in emissions. The remaining four have been used in

other fields (66). An application not mentioned in (66) is analyzing sensitivity coefficients to provide information about compensating errors. Good candidates are quantities that have sensitivity coefficients of nearly equal magnitudes and opposite sign.

Model sensitivity studies tend to be local parametric analyses that rely on the model being structurally correct. The range of applicability of sensitivity results is limited by the soundness and validity of the air quality modeling system and the supporting database employed to develop inputs. Although sensitivity analyses can be conducted using any model, the usefulness of such findings will be determined by how well the model actually represents key physical and chemical phenomena. Many sensitivity analyses have been conducted using models that do not provide a full representation of key atmospheric processes. Such studies using 0-D or 1-D models have limited scope and provide useful results within a restricted context, such as assessing the impact of uncertainties in chemical reaction rate constants on the calculated net ozone formation rate. The scaling of sensitivity results with model complexity is not understood. Neither are the limits of the usefulness of information derived from simple models for applications to more complex models. Considerable care must be exercised to determine the applicability of the findings in a regulatory regional air quality modeling context.

As mentioned previously, one application of sensitivity analysis relates the uncertainty in a quantity of interest, such as peak ozone concentration, to uncertainties in a suite of influential parameters. This type of analysis, referred to here as sensitivity/uncertainty analysis, most frequently estimates the uncertainty in model predictions resulting from uncertainties in inputs. Notable examples of the approach are reviewed herein. Before embarking on this discussion, however, attention is given to two important technical issues that have a significance for the validity and utility of sensitivity analysis results.

Air quality models are evaluated over a limited range of conditions, typically those associated with a set of historical meteorological events and their associated emissions conditions. Individual model components, such as the chemical mechanism, may be evaluated by independent means (e.g., smog chamber experiments) that cover a broader range of conditions. In designing sensitivity analyses and computer experiments that involve modification of model inputs, care must be exercised in (a) choosing the range over which parameters are evaluated and (b) evaluating the results in situations where the inputs are set to values that extend significantly beyond the range for which the model has been evaluated. Unfortunately, sensitivity studies are often conducted using models that have questionable or yet-to-be-determined capacity to replicate historical air quality events.

The second issue is correlation among the input variables. In selecting model input values, one must assure that all values are indeed plausible and consistent. Changes in correlated variables, such as emissions estimates and boundary conditions, may not be independent. Accurately representing such interdependencies among model inputs in the design of sensitivity experiments can (67–70) and should be achieved.

STRUCTURAL SENSITIVITY ANALYSIS Structural sensitivity analysis is concerned with characterizing the response of the model outputs to a change in the basic representation of physical and chemical processes or in the structure of the model itself. Analyses explore the influences that representations have on estimated pollutant concentrations. For example, several investigators examined differences among various chemical mechanisms developed for use in air quality modeling [for examples, see (71) and (72)]. Similarly, alternative means are available for representing plume rise, turbulent diffusive transport, and for solving the governing equations themselves. Each of these choices has some impact on the resulting calculated pollutant levels. Comparing results generated using models of different dimensionality (i.e., 0-D, 1-D, 2-D, or 3-D) offers insights about the description of transport [for example, see (73)].

Although it is relatively easy to design mathematical and statistical methods to explore parametric sensitivity, the treatment of structural sensitivity and its ultimate relation to uncertainty is more challenging. Analyses of this type have principally relied on model-to-model comparisons and expert judgment, which are discussed below.¹² As improved representations of atmospheric processes are developed and incorporated into air quality models, structural sensitivity studies will provide a means for describing how the improvements may alter the outcome of previous regulatory modeling assessments. They may be used to support claims that the improved model performs as well as or better than other models that EPA or CARB have approved for use in regulatory applications. When structural sensitivity studies are performed using 0-D or 1-D models to reduce computational burden, the results may provide an initial indication of the sensitivities that may be found in more complex 3-D models. Such structural analyses provide an indication of the advantages and limitations of using less complex models for diagnostic purposes.

Like parametric and functional sensitivity analyses, the utility of results from structural sensitivity analysis is critically dependent on the ultimate soundness and validity of the modeling constructs and supporting databases. Of particular concern are situations where the results are being used to support regulatory decision making.

Methods

The starting point for any sensitivity analysis is an air quality model with a prescribed set of inputs that provide the best representation of all significant atmospheric phenomena. There are several approaches for calculating sensitivity coefficients, each with advantages and limitations.

Local parametric sensitivity analysis is the most prevalent type of sensitivity analysis applied to air quality modeling. Tilden et al. (64) presented a comprehensive

¹²In the interest of parsimony, literature pertaining to uncertainties associated with the representation of turbulent transport and meteorological modeling and parameterization is not reviewed in detail. See the reviews by Seaman (44) and Russell & Dennis (32).

early review of sensitivity techniques, which included some applications to atmospheric phenomena. Hamby (74) recently reviewed sensitivity analyses techniques applied to air quality modeling.

The simplest and most frequently used sensitivity analysis is the perturbative approach. Often referred to as the brute force method (BFM), a parameter value is perturbed, the model is rerun, and the change in observables is calculated to determine sensitivity coefficients. If the perturbation is small enough, as dictated by the numerics and the actual sensitivity of the parameter of interest, a first-order sensitivity coefficient is obtained. This approach can be used to check more refined methods and algorithms. Using large perturbations along with another more formal method allows one to deduce the limits of various orders of sensitivity coefficients. The BFM approach can also be used to conduct global sensitivity analysis.

The perturbative approach does not work well when there are a large number of observables or parameters of interest. When conducted using a 3-D model involving many input parameters, the computational requirements can quickly become overwhelming. To ameliorate this situation, simpler 0-D and 1-D models are often used to explore specific sensitivity issues. Given the large number of variables subject to study in a 3-D model, statistical design procedures are often used. Both fractional and second-order design procedures may be employed to reduce the number of model simulations to a more manageable level while providing reasonably accurate estimates of the sensitivity coefficients. When several model simulations are undertaken, considerable knowledge about model sensitivity characteristics is obtained. Thus, it may be possible to use existing findings of model sensitivity characteristics to update the experimental design to achieve desired goals more efficiently.

There are a number of approaches in which sensitivity differential equations are derived from the original parent equations and solved along with them. The equations can be derived analytically, as in the decoupled direct method (DDM) (75–77), or by using automatic differentiation, like Automatic Differentiation of Fortran (ADIFOR) (78, 79). A distinction needs to be made here: Most DDM applications focused on first-order sensitivity equations. A separate set of equations has to be derived for higher-order sensitivity coefficients. The number of second-order equations increases as the square of the number of first-order equations. Many DDM applications have been concerned with simpler models (0-D and 1-D). Recent research by Yang et al. and Dunker et al. [see (18) and (77)] developed the DDM approach for a 3-D model to explore the sensitivities of pollutant concentrations to initial conditions, boundary conditions, and emissions. To enhance computational efficiency without sacrificing accuracy Dunker et al. (77) developed an algorithm by deriving sensitivity equations from the formulas of the hybrid chemistry solver and the nonlinear advection algorithm. The effort produced highly accurate first-order sensitivity coefficients.

The advantage of DDM is that it computes systematically all sensitivity coefficients for parameters of a certain type for all dependent variables. The disadvantage is that it is computationally intensive and can be prone to numerical errors when

approximations are made to enhance computational efficiency. Another disadvantage is that sensitivity coefficients calculated are almost always first order. When these coefficients are used in uncertainty calculations, parameter variations often exceed those for which the first-order approximation is valid.

The Green's function method (GFM) generates a Green's function (GF) or a propagator that can be used to compute all orders of sensitivity coefficients (80). For the ADE, the derived sensitivity equations are linear inhomogeneous equations with time-varying coefficients. The GF is the solution to the homogeneous equations derived from the sensitivity equations. If there are n observables, the GF dimension is $n \times n$ and most of the computational burden is involved with calculating it. If the number of parameters is large, the extra computational cost is only related to finding specific solutions for each parameter.

With efficiency comparable to DDM, the GFM offers several advantages. It provides sensitivities to initial conditions with no extra computational effort. It can be used to obtain higher-order sensitivities because sensitivity-governing equations of all orders have identical homogeneous parts. Consequently, the only extra computational effort for computing higher-order sensitivities is in finding their particular solutions by quadrature. Furthermore, the GF is an actual sensitivity. The GFM can be used to explore how a dependent variable at a given location and time in the modeling domain is sensitive to the value of another dependent variable at another location and time. Second-order sensitivities are sensitivities of sensitivity coefficients. As such, they are useful for exploring parametric interdependencies, because they indicate how the sensitivity of one parameter changes with a perturbation in another parameter.

The adjoint approach is related to the GFM. As a transpose of the GF, the adjoint operator is useful for sensitivity and inverse analysis. It is especially attractive for air quality modeling when the number of input parameters is large relative to dependent variables of interest. Adjoint methods have been used for data assimilation to improve emissions estimates that bring modeled and measured results into closer agreement [see (81)].

Scale requirements (i.e., local or global) also have an impact on the sensitivity analysis approach. As the scale increases, there is a potential need to treat higher-order derivatives and interaction terms that might otherwise not be as important in a local analysis situation. This is especially important for the most influential variables that tend to have the largest sensitivity coefficients. For them, perturbations in excess of 15% often require second- and higher-order analyses.

The Fourier amplitude sensitivity test (FAST) method of (82) and (65) is a nonrandom, efficient sampling procedure defined by determining a search curve in the space of the input parameters so that the path length included in a given portion of parameter space is proportional to the joint probability distribution. Compared to random sampling, the main advantage of FAST is that it is relatively easy to determine the minimum number of sampling points required for obtaining reliable sensitivity coefficients. Like stochastic sensitivity analysis, FAST assumes that the uncertainty in each parameter can be described by a known probability

distribution. It also assumes that all parametric variations are independent, which often is not the case.

Although sensitivity coefficients identify the most influential parameters and reveal how a system responds to changes, they do not indicate which parts of a model are most uncertain or assess the size of the uncertainty. Parameter uncertainties must be known or estimated prior to conducting uncertainty analysis with parametric sensitivity coefficients. Individual parameter uncertainties are best described by a probability distribution over the range of parameter uncertainty. Input values are sampled from the parameter probability distributions, and simulations are executed for each set of parameter values. In the majority of studies, the sampling is done in a random or quasi-random way, often using Monte Carlo techniques. The multiple simulations produce model output probability distributions that reflect the model uncertainty due to the parameter uncertainties. Frequently a constrained sampling scheme similar to Latin hypercube sampling, as developed by McKay et al. (83), is used to improve efficiency. Although sensitivity coefficients are not needed in these applications, statistical methods, such as multiple regression analysis, are often used to attribute the resulting uncertainty to the different input parameters. Doing so provides a measure of the sensitivity coefficients.

Tatang et al. (84) also developed an efficient approach for determining model uncertainty due to parameter uncertainty. They approximate a model response surface with orthogonal polynomials in the model parameters. The weighting functions of the polynomials are the probability density functions (PDFs) of the parameter uncertainty. The method is significantly faster than Monte Carlo approaches, but often the PDFs are not known.

An approach that appears quite attractive for sensitivity/uncertainty analysis is the use of Bayesian updating. This type of analysis incorporates actual observations, as best illustrated by example. Bergin & Milford (85) used Bayesian Monte Carlo analysis to refine the uncertainty estimates based on their previous work by using observations to update the uncertainty estimates. With a standard sampling technique, values are sampled according to the parameter a priori probability distributions, and each simulation is given an equal weight. When using Bayesian updating, observations are used to compute a likelihood function and a corresponding weight for each simulation. The weights are used to update a posteriori the uncertainty estimates for the model output and input parameters. Bayesian methods potentially allow for evaluations of the parameter uncertainty estimate by using additional information from pollutant concentration measurements. They may also be useful for comparing models that are structurally different, thus aiding in the difficult task of assessing structural uncertainty.

There are, however, important challenges associated with using Bayesian methods. Construction of the likelihood function requires a good understanding of the uncertainty in observations. Without this description, results may be unreliable. Furthermore, current techniques assume that variables used for updating have uncorrelated uncertainties. This is an important limitation when more than one observation variable is used (e.g., consecutive measurements of one

concentration, concentrations of different species, or at different locations). At this point, the errors in observations may not be understood well enough to rely on Bayesian techniques for air quality modeling.

A closing note—sampling-based parametric sensitivity studies involve planning a set of simulations and conducting them in succession. The n^{th} simulation in a set of p simulations is selected and designed based on what is known before any of these simulations is conducted. In contrast, Bayesian design involves planning the n^{th} simulation using what is known at the beginning and what is learned through the $(n - 1)^{\text{st}}$ simulation. (In fact, this is what air quality modelers tend to do subjectively.) Thus, the choice of the n^{th} simulation can be better informed and the set of simulations more efficient. In short, there is experience with less efficient parametric planning, but potentially more effective Bayesian methods are not well developed and are difficult to apply. Clearly, the development of practical and effective sequential planning techniques would be most welcome.

Applications

Selected sensitivity and sensitivity/uncertainty analyses are listed in Table 2.

Summary

Having reviewed research associated with conducting sensitivity and sensitivity/uncertainty analyses, several key observations emerge:

- The ultimate utility of sensitivity results is dependent on the soundness and validity of the model and supporting database. Care must be exercised to ensure that sensitivity results are used within their range of applicability.
- Few of the approaches demonstrated thus far have been used to contribute to model validation or diagnosis of sources of errors. There are promising new techniques for 3-D models.
- The usefulness and relationship of sensitivities calculated for 0-D and 1-D models to those associated with 3-D models is not well understood and warrants further investigation.
- With a few exceptions, second-order sensitivities estimation is generally lacking, but is especially useful for exploring interdependencies among inputs.
- Sampling requirements scale with input parameters; however, there is encouraging effort being directed toward the calculation of sampling needs and efficient, formal sampling techniques.
- Bayesian updating is a promising approach, yet remains limited by the need to understand a priori uncertainties and correlations associated with input parameters and observations used for updating. Nevertheless, in principle, sequential planning (i.e., Bayesian) techniques are inherently more efficient and, therefore, more attractive than simultaneous (i.e., parametric) planning techniques.

TABLE 2 Selected sensitivity studies using PAQSM

Method	Dimensionality	Types of analyses performed	References
Brute force method (BFM)	0-D ^a and 3-D ^b	Computed O ₃ sensitivity to effects of motor vehicle emissions inventory updates and choice of chemical mechanism.	(25)
Direct decoupled method (DDM)	0-D, chemical mechanism of Regional Acid Deposition Model version 2 (RADM2)	Computed sensitivity of O ₃ , HCHO, HNO ₃ , peroxyacyl nitrates (PAN), and H ₂ O ₂ to 157 reaction rate parameters and stoichiometric coefficients of 34 reactions using a regional-scale gas-phase chemical mechanism. Sensitivity coefficients are combined with uncertainty estimates of the chemical mechanism input parameters to evaluate contributions to modeled pollutant concentration uncertainties.	(87)
Latin hypercube sampling (LHS) and multiple linear regression	0-D, chemical mechanism of RADM2	Uncertainties in estimated ozone reductions for three control strategies (reducing NO _x or VOC emissions by 25% or both by 25%) are computed under six different VOC-to-NO _x ratios with random sampling using a LHS/Monte Carlo technique. Multiple linear regression analysis is used to identify the most significant input parameter uncertainties.	(88)
LHS and multiple linear regression	1-D, ^c trajectory version of Carnegie/California Institute of Technology airshed model (CIT)	Computed the influence of uncertainties of input parameters (29 reaction rates, deposition affinities, emissions, wind direction, mixing height, and atmospheric stability) on uncertainty in estimated ozone concentrations. LHS is used to compute probability distributions for estimated concentrations of O ₃ , HCHO, HNO ₃ , PAN, and NO _y . Multiple linear regression analysis is used to attribute secondary pollutant concentration uncertainties to the input parameter uncertainties. Furthermore, peak ozone concentration uncertainties are estimated under two scenarios (25% motor vehicle NO _x or VOC emissions reductions).	(89)
DDM	0-D	Computed O ₃ and PAN sensitivity to reaction rates and initial conditions.	(90)
DDM and LHS	0-D	Determined influence of reaction rate uncertainties on VOC reactivity.	(91)
DDM and LHS	0-D	Computed the influence of reaction rate uncertainties and alternative fuel exhaust variability on fuel reactivity adjustment factors, while considering product yields.	(92)
DDM	3-D CIT	Calculated ozone sensitivity to its initial concentration and deposition rate, selected reaction rates, precursor concentrations, and wind speeds.	(18)
Fourier amplitude sensitivity test (FAST)	0-D	Determined sensitivity and uncertainty due to reaction rates.	(93)
BFM	3-D	Investigated effects of NO _x and VOC controls on reducing O ₃ , PAN, and NO ₃ .	(14)

(Continued)

TABLE 2 (Continued)

Method	Dimensionality	Type of analyses performed	References
BFM	3-D	Explored using sum of total reactive nitrogen (NO _x) as a metric for determining ozone response to various reduction strategies.	(15)
BFM	3-D	Explored response of O ₃ and its precursors to different emissions reduction strategies.	(94)
BFM	3-D	Estimated influence of reaction rate uncertainties on VOC reactivity due to uncertainty in 14 reaction rate parameters identified as the most influential by Yang et al. (91).	(17)
Monte Carlo (MC) with linear optimization	Uses optimization model	Used a cost-benefit framework to evaluate the use of VOC reactivity to identify reduction priorities to regulate urban O ₃ given uncertainties in emissions and VOC reactivity parameters.	(95)
MC with subjective judgment analysis	3-D	Computed uncertainty in estimated O ₃ concentrations due to uncertainties in over 100 input parameters that were estimated through elicitation of expert opinions.	(19, 20)
Green's function	0-D	Identified the most influential chemical reactions in photochemical smog production in terms of the sensitivities of pollutant concentrations with respect to rate constants and the sensitivities of one pollutant concentration at a specified time to concentrations of other pollutants at previous times. In addition, computed second-order sensitivity coefficients for exploring interdependencies between input parameters.	(80)
MC with Bayesian updating	1-D trajectory version of CIT	Used observations to modify uncertainties in input parameters, refined results of (89).	(85)
DDM	0-D	Compared alternative treatments of the organic chemistry kinetics using a formal gradient-based approach to compare three atmospheric chemical mechanisms. Computed sensitivities for O ₃ , H ₂ O ₂ , and HCHO.	(71)
Structural	0-D to 3-D	Summarized several modeling studies to compare urban ozone control. Examined uncertainty issues that arise when using VOC reactivity for ozone control.	(73)
DDM, BFM, and LHS	1-D and 3-D	Compared VOC reactivity changes determined in two models [3-D grid model and trajectory model used by Carter (96)] due to changes in VOC emission estimates. The emissions were changed by a fixed fraction, and the effect was determined using three metrics: peak O ₃ , population exposure, and spatial exposure.	(16)
Functional analysis	3-D	Functional sensitivities of an objective function were derived from sensitivity densities. Functional analysis was used to determine source-receptor relationships for acid deposition.	(63)

^a0-D = single box, zero-dimensional model.

^b3-D = gridded, three-dimensional model.

^c1-D = column trajectory, one-dimensional model.

- Experience and methods are limited with respect to promising structural uncertainty assessment. Efforts to date have involved model-to-model comparisons and subjective analysis. Better theoretical understanding of structural sensitivity analysis in air quality modeling would greatly improve the reliability of models.
- Further work is needed to establish practical methods for using pertinent sensitivity/uncertainty analysis techniques to develop estimates of uncertainties associated with 3-D regional photochemical modeling results.

Although this summary highlights limitations associated with sensitivity analysis, such analyses are often the most expedient available. When conducted using a model that has undergone careful evaluation to identify and, where possible, to reduce or eliminate bias, sensitivity analysis may offer the best opportunity for understanding and estimating uncertainty.

V. DIAGNOSTIC ANALYSES

Overview

Diagnostic analyses are an important component of a comprehensive evaluation of air quality model performance (97, 98) and are carried out for two main purposes. When conducted in a prescriptive (or defined) manner, they provide insight into how well the modeling system represents particular physical or chemical reaction phenomena. Positive outcomes of such assessments help to bolster confidence in the modeling results. Equally important, indications that the modeling system is not simulating important phenomena adequately can help to direct subsequent efforts associated with the second type of diagnostic analyses aimed at rectifying model performance problems. In this case, the specific analyses undertaken depend on the nature of the modeling problem that has been encountered and the creativeness of the investigators in identifying the possible underlying causes based on the symptoms exhibited in the modeling results.

Diagnostic analyses evaluate modeled representations of particular phenomena that are known or thought to play key roles in influencing pollutant concentrations. Based on prevailing understanding and available measurements, specific analyses are designed and carried out to provide a quantitative determination of model performance in relation to the phenomena of concern. Analyses may involve the comparison of model variables, such as wind speeds and directions, temperatures, and ratios of selected pollutants with pertinent observations.

Diagnostic analyses should only be used to help point the way to modified treatments of atmospheric processes or model inputs based on sound scientific facts and representations. Particular care must be exercised in using such results to tune the model. In general, model tuning should be avoided. Scientific principals should determine how best to represent atmospheric processes and to develop model inputs.

Limitations of Available Measurement Data

Diagnostic analyses provide direct quantitative information concerning the representation of particular atmospheric phenomena in a PAQSM. The ability to assess the quality of representations is determined by the availability of pertinent measurements. The more suitable the available measurements, the more quantitative and substantive the assessment. If measurements are very limited or unavailable, then the assessments must of necessity be qualitative and based on the general knowledge of the investigator.

The data needed to support diagnostic analyses are generally only available through the conduct of special field measurement studies. Quantitative analyses will only be possible when such measurements are available. Routine measurement programs provide basic observations of surface pollutant concentrations and meteorological variables (such as wind velocity, temperature, and humidity) and are intended to fulfill regulatory requirements. They are not designed to provide information needed typically to support diagnostic analyses. Of particular concern is the general absence of air quality and meteorological data aloft. Detailed emissions data are rarely available with which to assess issues concerning the adequacy of the emissions inventory. Similarly, observations of dry deposition are rarely available.

When pertinent measurements are not available, analyses are formulated and assessed based on the general understanding of what may or may not be a reasonable representation of the phenomena of interest. For example, Jang et al. (21) applied process analysis techniques to obtain a better understanding of the high resolution version of the Regional Acid Deposition Model (HR-RADM) model results in the New York area. Although available pertinent measurements were limited, they were able to identify questionable behavior in the calculated pollutant concentrations.

Examples of Diagnostic Analyses

Four studies noted herein illustrate existing approaches for conducting diagnostic analyses. Jang et al. (21) applied process analysis techniques to investigate the effects of grid resolution on ozone formation chemistry. Chemical process analysis was used by Tonnesen & Wang (99) to examine a PAQSM applied to the Houston area. Roth et al. (100) and Vuilleumier et al. (101) provided examples of the types of diagnostic analyses that might be performed given the availability of extensive field measurements. And finally, a number of innovative analyses have been carried out in support of regulatory modeling in the northeastern United States (105–109).

PROCESS ANALYSIS TECHNIQUES Jang et al. (21) studied the influence of grid resolution on calculated ozone levels in the New York area using the high-resolution version of a regional acid deposition model, HR-RADM. This model included provisions for producing detailed information on the contributions of key emissions, transport, and chemical reaction processes to calculated ozone

levels. The technique is termed the integrated reaction rate/mass balance (IRR/MB) method, originally developed by Jeffries (102) to study chemical mechanisms. Details of the derivation are provided by Jeffries & Tonnesen (38). The technique was implemented in a trajectory model (103).

HR-RADM simulations for the northeastern United States were conducted for a multi-day ozone episode that occurred during the summer of 1988 using horizontal grid resolutions of 80, 40, and 20 km. The investigators noted peak ozone predictions near New York City that were 0.104 and 0.139 ppm using 80 km and 20 km grid resolutions, respectively. The IRR/MB method was used to investigate the ozone production processes at the two grid resolutions. The investigators were able to determine key differences between the 80 km and 20 km grid cases. They found that the interactions of chemistry and vertical transport, both competing for the emitted NO, was the cause of differences in results. This suggested that to improve model accuracy in calculating ozone formation, it was necessary to have adequate horizontal and vertical grid resolution and that the representation of the vertical transport process in the model needed a timescale corresponding to the relatively fast chemical reactions.

Tonnesen & Wang (99) discuss a more recent application of chemical process analysis to photochemical model simulations carried out in the Houston area. They conducted analyses to provide new insights into the chemical processes that affect ozone formation and to identify important sources for uncertainty. In particular, their analyses raised questions concerning the large fraction of NO_x emissions that were converted to inert HNO₃ during the nighttime N₂O₅ chemistry. There is a large uncertainty associated with the rate of this reaction, which in turn may have a significant influence on the responsiveness of the modeled ozone estimates to NO_x emissions changes. This could be quite important given the possible implementation of NO_x emission controls in the Houston area.

TECHNIQUES USING EXTENSIVE FIELD MEASUREMENTS Extensive measurements were conducted in central California in the summer of 1990 as part of the SJVAQS/AUSPEX field measurement program to support a PAQSM application. DaMassa et al. (104) discussed the results of an operational evaluation of the modeling application. The SJVAQS/AUSPEX program provided a wealth of data with which to conduct diagnostic analyses of the meteorological, emissions, and air quality components of the modeling system.

Roth et al. (100) discussed the specification of process-related tests for a PAQSM. By constructing tests focusing on individual atmospheric processes, one can establish the accuracy of representation of these processes. If a particular model use is emphasized and requirements for accuracy of estimation are prescribed, "thresholds for acceptability of performance" of individual modules can be established through sensitivity analysis. Roth et al. (100) also introduced the concept of *thresholds triggering concern*, which connotes specified levels of bias in estimation which, if exceeded in performance, indicate the need for diagnosis of flaws and improvement in performance before the model can be tentatively accepted

for use. A *stressfulness index* can be defined that describes the degree to which a model is forced to reveal flaws. The index provides a quantitative, albeit heuristic, measure of the degree of challenge offered a model through a suite of tests.

Roth et al. (100) developed a set of 24 questions for assessing the SJVAQS/AUSPEX Regional Mapping of Air Pollution (SARMAP) modeling system applied to the San Joaquin Valley, California. Two examples of these questions include:

- How well does the model simulate eddy circulation in the central and southern portions of the valley?
- Are ambient and emissions ratios of hydrocarbons to NO_x and CO to NO_x concentrations consistent?

Specific process-related tests were then derived to provide answers to each question. For each question, Roth et al. (100) provided commentary concerning the importance of the question, as well as a statement of the test, a specified threshold triggering concern, a brief discussion of the availability of data needed to support the proposed test, and an assessment of the feasibility of conducting a meaningful test.

Vuilleumier et al. (101) investigated sources of uncertainty in estimated photolysis reaction rates of NO_2 through comparisons with actual measured values from a chemical actinometer operated as part of the 1997 Southern California Ozone Study. The aerosol single scattering albedo was found to be a major source of uncertainty in the photolysis estimates, yielding biases between calculated and measured values ranging from 17%–36%.

A considerable body of analysis has been carried out to support regulatory modeling in the northeastern United States. For example, Hogrefe et al. (105–107) and Biswas et al. (108) demonstrate the application of spectral decomposition techniques to assess the ability of regional meteorological and air quality models to adequately represent observed intraday, diurnal, synoptic, and longer-term fluctuations in meteorological variables and primary and secondary pollutant concentrations. Using ambient pollutant measurements collected at the surface and by aircraft, Zhang et al. (109) examined the influence of high ozone trapped aloft at night on the temporal evolution as well as the peak ozone levels observed near the surface during the day.

Future Needs

To date, there is no formal guidance on how to conduct diagnostic analyses in a systematic manner. This may be reflective of the larger issue of the adequacy of aerometric databases available to support modeling. Until commitments to secure such databases are made, the possibilities for quantitative diagnostic analyses will be limited. Nevertheless, the application of process analysis to provide a general assessment of the representation of atmospheric processes in the modeling system is a viable alternative. Further effort is needed to develop guidelines for the implementation of such an approach and for the interpretation of the results.

Process-related tests employing available aerometric and emissions data provide another diagnostic assessment pathway. This approach is of particular value in cases for which a supplemental field measurement program has been implemented to provide a better characterization of key atmospheric processes. A design for such a testing program has been developed for model applications in central California. Actual implementation of the program is needed to assess the utility of the results and to provide guidance in how best to formulate extensions for application to other regions.

VI. CORROBORATIVE/ALTERNATIVE MODELING AND SUBJECTIVE JUDGMENT

Overview

Corroborative modeling analyses can play an important role in a comprehensive assessment of model performance by providing an independent means of assessing some aspect of the air quality model results, such as the likely influence of reducing emissions of VOCs or NO_x on ozone. Consistency of findings enhances confidence in the modeling results. An alternative modeling system might be applied that has been demonstrated to provide sound results in previous studies. The alternative base case approach provides a lower bound estimate of uncertainties in modeling results by using alternative sets of model inputs, each within its range of uncertainty. Finally, subjective judgment may be applied wherein the knowledge and experience of one or more individuals is employed to render an estimate of the uncertainty.

The utility of the information developed in the various corroborative approaches will depend on the methodology pursued and its applicability to the question of interest. Each approach to be considered must be evaluated a priori as to its merits. Yet, whatever the results, each will provide only partial information about the uncertainties of interest.

Subjective Judgment

Early efforts to query human judgments to support decision making and policy formulation resulted in the Delphi method (110, 111). Views of experts were solicited, summarized, and circulated in summary form among the same experts. The objective was to identify, resolve, and document differences—and iterate to a view that could be supported by all or most participants. In doing so, a fundamental constraint was to avoid creating an average or typical view.

Morgan & Henrion (9) recommended that expert elicitation be used to estimate uncertainties when needed, such as input parameter probability distributions for use in sensitivity/uncertainty simulations. Hanna et al. (20) pursued this course to evaluate uncertainties associated with modeling to identify emissions controls needed to attain the NAAQS for ozone in the eastern United States. As with most subjective methods, the following limitations pertain:

- Experts may differ substantially in their estimates, and those differences may be irresolvable due to insufficient available data.
- Experts have been and might be overconfident about the accuracy of their estimates of uncertainty.
- The estimates developed may be difficult to use.
- Because of questions surrounding their reliability, subjective uncertainty estimates may be disregarded in practice.

Nevertheless, expert judgment may be a welcome method for uncertainty estimation in the absence of feasible and reliable alternatives.

Corroborative Analyses

Corroborative analyses can provide an independent means of model evaluation (97, 98). Data analysis studies are an important source of information for corroborating model treatments of atmospheric processes and model results. Such studies can provide independent insights into the characteristics of key atmospheric phenomena. With such information, tests can be devised to assess the adequacy of model performance.

Another important avenue for corroborative analysis involves the application of observation-based methods. Such methods have been developed to determine if, for a region or subregion, VOC or NO_x controls are likely to be most effective in reducing ozone concentrations. For example, Blanchard et al. (112) use NO , NO_y , and O_3 data collected at the same location as input to a simple algebraic model described in Johnson & Azzi (113) to determine if VOC or NO_x control is preferable. Example applications of this procedure are provided in Blanchard et al. (114–116).

Developed by the Aeronomy Laboratory of the National Oceanic and Atmospheric Administration (117), a second approach relies on the use of a correlation between peak ozone and NO_y concentration. If an area is NO_x limited, NO_y and peak O_3 should correlate. If the area is VOC limited, they should not. Both procedures rely on the use of NO_y and O_3 data to make the necessary calculations, depend on data analysis alone, and bypass the need for modeling and compilation of an emissions estimate. However, accurate measurement of NO_y is quite difficult.

Cardelino & Chameides (118) use an observation-based model to describe O_3 production in terms of its precursors by employing the concept of relative incremental reactivity. Box model calculations are made to determine the amount of O_3 produced in daylight hours at specific sites, as well as the sensitivity of O_3 production to changes in precursor levels. An incremental reactivity is defined in terms of the O_3 -forming potential difference for an assumed incremental change in the ratio of the precursor concentration and the integrated amount of precursor emitted or transported to each site. The method identifies four regimes of O_3 formation that range from NO_x limitation to VOC limitation and segregate the VOC into natural and anthropogenic contributions. Extensive discussions of the

application of several contemporary observation-based analysis approaches are provided by Blanchard, Kleinman, and Trainer et al. (119–121).

Alternative modeling approaches may also be adopted in a corroborative manner. For example, an alternative prognostic meteorological model might be employed to develop meteorological inputs. Or, simulations might be conducted using a second air quality model that provides somewhat different treatments of one or more atmospheric processes. A good example of such an approach is being carried out to support SIP-related activities in the Houston area. Meteorological fields have been developed using three different meteorological models: Systems Applications International Mesoscale Meteorological Model (SAIMM), National Center for Atmospheric Research/Pennsylvania State University Meteorology Model (MM5), and Regional Atmospheric Modeling System (RAMS). In addition, air quality simulations have been conducted using two air quality models: Comprehensive Air Quality Model extended version (CAMx) and Community Multiscale Air Quality Model (CMAQ). Motivation for such efforts is related to the difficulties encountered in accurately simulating occurrences of high ozone concentrations. The ability to adequately simulate the complex meteorological phenomena that occur in the area continues to be a challenge. In addition, there is some evidence that highly reactive VOC emissions may be underestimated. Thus, efforts were undertaken to develop suitable adjustments to the estimate as an interim measure until more accurate emissions data are obtained. Documentation prepared by the Texas Commission on Environmental Quality discusses the various alternative modeling efforts (122).

A similar analysis for the northeastern United States using both MM5 and RAMS meteorological fields as inputs to the Variable Grid Urban Airshed Model (UAM-V) model is discussed by Biswas & Rao (123). While neither modeling system provided significantly better ozone performance, model-to-model and episode-to-episode differences were noted in individual grid cells with regard to the impacts of precursor emission reductions on calculated ozone levels.

Another corroborative approach is the concept of alternative base case (ABC) analysis (124, 125). The central premise of ABC analysis is that there are many combinations of model inputs that will produce model estimates—ozone concentrations as a function of location and time—that are similar and, in fact, are indistinguishable in accuracy within the error bounds accompanying the estimates. Possible combinations of alternative input settings are derived from an assessment of the uncertainties in each of the key input variables. Consideration is also given to the manner in which inputs were developed, particularly possible sources of compensating errors. The elements of ABC and sensitivity/uncertainty analysis (discussed in Section IV) have some common traits.

In a hypothetical and simple case of two sets of conditions [(a) VOC emissions, E_1 , and VOC boundary conditions, B_1 , and (b) VOC emissions, E_2 , and VOC boundary conditions, B_2 , and all other conditions the same as for case 1], estimated ozone concentrations as a function of time may be quite similar, with differences of the order of estimated measurement uncertainties. This situation

can arise when the total loading of VOCs (initial conditions, boundary conditions, and emissions) is about the same magnitude, with similar spatial and temporal distributions for both cases. In practical applications, it may be possible to identify several sets of conditions that produce like outputs. Each may merit being accorded the status of base or reference case; selection becomes arbitrary. ABC analysis encourages identification of as many alternative base cases as possible. If five cases emerge from analysis, then each can be used as a reference case for examining emissions control strategies. Five results will be generated, and their spread in magnitude constitutes a lower bound on uncertainty in estimation of future ozone concentrations. If another ABC were to be identified, it can only increase (or maintain) the spread in outcomes. ABC analysis can be quite powerful, particularly in demonstrating the range of potential future outcomes for alternative model input conditions that are indistinguishable for a given data base.

To carry out such assessments, it is necessary to identify one or more suitable corroborative analysis approaches, such as those described above. Then a pertinent set of PAQSM results must be obtained that can be related to those produced by the selected analysis approach(es). After the appropriate analyses and model simulations are carried out, the consistency of the results must be determined. Further diagnostic and evaluation studies may be needed if the PAQSM results are not corroborated by the independent analyses.

Potential Uses Within Current Regulatory Context

Corroborative analysis approaches provide supplemental information that may confirm the findings of PAQSMs. In this context, they may contribute to weight of evidence arguments while helping to build confidence in findings. Equally important, the supplemental information may raise questions that motivate further diagnostic assessments before PAQSM findings are confirmed.

The utility of information derived from subjective judgment is dependent on the relevance of the experience of the participants. The information may be quite germane, or it may be incorrect and misleading.

Observation-based approaches can provide information within the range of circumstances represented by the available observation set. However, extrapolation beyond the range of existing conditions will involve considerable uncertainty. Consequently, observation-based methods have limited ability to estimate accurately the effects of emissions controls that significantly alter precursor emissions. They may be best suited for confirming PAQSM indications of the general directional effects of VOC and/or NO_x controls on ozone levels.

Application of sophisticated alternative modeling approaches can provide quantitative information about modeling uncertainties, especially when interpreted in the context of alternative base case analysis. However, if some aspect of the model is flawed, such as the meteorological or emissions inputs, then the resulting information will be of little value and possibly misleading.

Over the past several years, considerable effort has been devoted to developing and applying various corroborative analysis approaches. Although existing regulatory modeling guidance recognizes the potential value of such approaches, further effort is needed to clarify the range of applicability of each approach and to indicate how such information can be related to quantitative estimates of uncertainties associated with PAQSM results.

VII. LOOKING TO THE FUTURE

Several observations derive from review of the literature and knowledge of applications of photochemical models in air quality analysis.

1. Uncertainties pervade the use of models. Consequently, a range of model estimates may be anticipated for a given set of inputs and their associated uncertainties. Thus, estimates of uncertainties should be made and factored into processes involving model-based decision-making associated with air quality issues.
2. Methods available today that are truly useful either partially address the estimation need or focus on a defined, limited part of the problem. Unfortunately, no method is now available for estimating uncertainty in modeling that is comprehensive in scope.
3. A comprehensive method for analyzing uncertainty information would (a) propagate uncertainty from each component of the modeling system through the system into an estimate of uncertainty associated with model output, (b) elucidate bias, and (c) account for variability. The method would also synthesize and integrate results from the various methods employed to estimate uncertainty comprehensively. The products of a comprehensive uncertainty assessment would be distributions or probabilistic statements characterizing the uncertainty of model estimates.
4. Developing a comprehensive approach to uncertainty analysis would be very valuable. Its feasibility should be assessed. Such an approach may not be possible, because nothing in the literature suggests so.
5. In practical applications, visual inspection of plots of concentration versus time for pollutant species of interest provides adequate information to determine if model performance is sufficiently acceptable to merit proceeding with comprehensive uncertainty assessment. In many cases, model performance is wanting. Where performance is unacceptable, major flaws in the model should be corrected prior to obtaining uncertainty information.
6. Sensitivity/uncertainty analysis is by far the most frequently used method for estimating uncertainty. Its focus is on the response of dependent variables to changes in inputs. When the response is significant, uncertainty is likely to be important; when the response is small, the converse is expected.

Sensitivity analysis does not address bias as an element in uncertainty explicitly. Generally, it is not suited for this use, because the main assumption made is that the model is substantially correct in its representation of reality.

7. As the key element of a comprehensive assessment of uncertainty, the development and application of methods for identifying, estimating, and reducing biases (i.e., mitigating or eliminating flaws in model representations) should be made a priority. This includes determining when bias is present, how to identify it, and what to do when it corrupts modeling results. Through assessment of bias, model formulation may be improved to increase the probability that the model is performing acceptably well for the right reasons and that modeled sensitivities are reliable. Examining the issue of potential bias typically requires case-specific procedures.
8. In addition to the evaluation of bias, natural and human-induced variability should receive attention in the comprehensive estimation of uncertainty. Some deterministic modeling formulations may simulate well-characterized stochastic processes using statistical sampling techniques, whereas others, such as those used to derive meteorological inputs, are incapable of simulating stochastic processes. The appropriateness and feasibility of developing stochastic models merits attention because they are potentially attractive means for incorporating variability.
9. Designing a comprehensive approach to uncertainty assessment that can be implemented and that addresses bias and variability requires a major research effort. To date, no such effort has been formulated, let alone undertaken. Rather, the focus has been on portions of the problem, in the absence of a more encompassing plan that might foster a more integrated research program design.

ACKNOWLEDGMENT

This research was supported by the Assistant Secretary for Fossil Energy, Office of Natural Gas and Petroleum Technology, through the National Petroleum Technology Office under U.S. Department of Energy Contract No. DE-AC03-76SF00098.

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