PAST, PRESENT, AND FUTURE AIR QUALITY MODELING AND ITS APPLICATIONS IN THE UNITED STATES

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Since the inception of the Clean Air Act (CAA) in 1969, atmospheric models have been used to assess source-receptor relationships for sulfur dioxide (SO_2), CO, and total suspended particulate matter (TSP) in the urban areas. The focus through the 1970's has been on the Gaussian dispersion models for non-reactive pollutants. The 1977 Amendments to the CAA mandated the use of dispersion models for assessing compliance with the relevant National Ambient Air Quality Standards (NAAQS) when new sources of pollution are permitted and for prevention of significant deterioration. In the 1980's, the focus has been on the secondary pollutants (e.g., ozone, acid rain), which led to the development of grid-based photochemical models to better understand the urban and regional scale pollution. In the 1990's, attention was paid to the development of one-atmosphere models to deal with multiple pollutants. The new NAAQS for ozone and fine particulate matter (PM2.5) that were promulgated in 1997 call for the use of one-atmosphere models in designing multi-pollutant emission control strategies. In the 2000's, there is considerable interest in the development of integrated airshed-watershed models to properly assess the effects of atmospheric pollution on sensitive ecosystems. Air quality models can help improve our understanding of the transport and fate of pollutants, and are essential tools for designing meaningful and effective emission control strategies. Future applications of air quality models will be towards the prediction and improved understanding of human exposure, especially in urban areas, and intercontinental-cross oceanic hemispheric air pollutant transport.

Air Quality Models

There are six types of atmospheric transport and diffusion models: Plume, Segmented Plume and Puff, Lagrangian Particle, Box, Eulerian Grid, and Computational Fluid Dynamics. Plume models assume conditions are horizontally homogeneous (everywhere the same) and steady state. Segmented Plume and Puff models divide the emissions into a series of overlapping volumes (or puffs) so that we no longer need assume horizontal homogeneous conditions or require conditions to be steady state. Lagrangian Particle models divide the emissions into tiny masses that are individually "traced" as they are stochastically transported and diffused downwind. The emitted gaseous or particulate material is "moved" at each time step by pseudo-velocities that take into account the three basic components of transport and diffusion: (1) the transport due to the mean wind, (2) the turbulent diffusion caused by the (seemingly) random fluctuations of wind components (both horizontal and vertical), and (3) the molecular diffusion (if not negligible). Box models assume the modeling domain is one large homogeneous volume (box); emissions entering this volume are assumed to be uniformly and instantaneously mixed throughout the volume. The top of the box may rise to simulate the rise of the mixing depth after sunrise, and pollutants aloft of this rising lid would then be entrained into the volume. The location of the box can be stationary (to simulate the air over a city), or can move with the transport wind (to simulate the 'aging' of an air mass). Eulerian Grid models divide the world into a three-dimensional array of rectangular cells ("grids") within which mixing is considered uniform and instantaneous. Grid models are used to simulate the formation of products through atmospheric chemistry and the removal of products by clouds and precipitation, all of which are usually sufficiently removed from the emissions of immediate concern that the "well-mixed" assumption in each cell is reasonable. Computational Fluid Dynamics (CFD) modeling is the science of determining a numerical solution to the governing equations of fluid flow whilst advancing the solution through space or time to obtain a numerical description of the complete flow field of interest. The governing equations for Newtonian fluid dynamics, the unsteady Navier-Stokes equations, have been known for over a century. However, the analytical investigation of reduced forms of these equations is still an active area of research as is the problem of turbulent closure for the Reynolds-averaged form of the equations. For non-Newtonian fluid dynamics, the theoretical development of chemically reacting flows and multiphase flows is at a less advanced stage.

Of the transport and diffusion models listed, plume and puff models are used operationally for air quality assessments of chemical species whose chemical/radioactive transformations can be represented using time-dependent decay approximations (e.g., SO_2, CO, primary emissions of particulate matter). Plume models are used for the near-field assessments and puff models are used for the meso-scale assessments. Eulerian grid models are used for operational air quality assessments of multi-pollutants having nonlinear chemical/radioactive reactions and secondary species of interest resulting from these reactions (e.g., ozone, sulfate and nitrate aerosols). Box models are primarily used for testing and comparison of alternative chemical kinetics. Regardless of the transport and diffusion model chosen, there are four additional fundamental requirements: characterization of the meteorological conditions, characterization of the emissions, characterization of the chemical/radioactive transformations, and characterization of the wet and dry deposition processes.

Meteorological characterization for air quality modeling has evolved considerably over the last three decades. Initially, fairly straightforward objective analyses of ambient surface and upper air data from routine meteorological monitoring networks was used to obtain gridded fields of winds, temperature, and moisture for input to Eulerian grid models, such as the Urban Airshed Model (UAM). This technique was facilitated by the ready accessibility of data from U.S. and Canadian centralized data sources. Limitations included the sparseness of data in key areas (e.g., mountain areas, seacoasts), interpolation and extrapolation uncertainties, and physical inconsistencies in the analyses, such as in the derivation of vertical wind velocities or boundary layer depths. The next methodological advance included physical constraints with the objective analyses of data. These included divergence minimization schemes to constrain mass fluxes and vertical velocities. These techniques were called diagnostic analyses of meteorological data, to distinguish them from the earlier objective analyses. Some of the same limitations (data sparseness, interpolation errors) applied to the diagnostic techniques as well. A major advance occurred in the mid-1980s when the Regional Acid Deposition Model (RADM) was linked to a prognostic meteorological model, the Penn State/NCAR Mesoscale Model (MM4). For the first time, fully self-consistent dynamically balanced meteorological fields were used to drive an air quality model. With the addition of four-dimensional data assimilation of observed profile data to the MM4 a few years later, error propagation and growth was controlled in the numerical meteorological simulation. The state-of-science since then has evolved with nested versions of mesoscale models (e.g., MM5, RAMS) with advanced data assimilation techniques as drivers for air quality models.

Emissions are an essential part of an air quality modeling application. Emissions have evolved from simple time-invariant estimates of pollutant mass to emissions modeling systems, which are as complex as the underlying chemical-transport model. For plume modeling, proper depiction of the relative position, size and amount of the release with respect to nearby receptors is critical. Early Gaussian dispersion model applications rarely considered temporal changes in emissions, although the Tennessee Valley Authority employed emission models in the mid-1970s for its supplementary control program that considered variations in power load, which were directly related to projected variations in stack parameters and hourly sulfur dioxide emissions. With the advent of photochemical models in the 1980s, emission estimates began to consider temporal variations (time-of-day and day-of-week) especially for mobile sources. The MOBILE models, first introduced in 1978, considered fleet composition, spatial variations, driving cycles, and temperature effects on evaporative emissions and cold starts. By the late 1980s, the important of biogenic hydrocarbons was recognized for photochemical models, as emission models such as the Biogenic Emissions Inventory System (BEIS) began to account for spatial vegetation patterns, temperature, solar radiation, and leaf phenology. Another important advancement in the late 1990s was the development of modeling systems and frameworks in handling the disparate emission inventories, emissions modeling systems, and model formats. For air quality management applications, projected emissions are a key consideration. Projections must consider the current state of emissions and using economic forecasts, anticipated technological changes, and future forecasts of land use and population patterns. Projected inventories and emission control strategies lay at the heart of air quality model simulation; they influence the confidence that can be placed in regard to the selected emission control strategy's efficacy in achieving compliance with the relevant NAAQS.

Photochemical kinetic mechanisms contain the set of reactions responsible for the generation of ambient ozone and other photooxidants, and are key components of air quality simulation models for ozone

and other chemically reactive ambient trace gases. Both inorganic (mainly NOx and HOx) and organic (HC) chemistry of the atmosphere are described in these mechanisms. Because the organic chemistry involves hundreds, if not thousands, of chemical reactions among several hundred chemical trace gases, mechanism compression techniques have evolved over time that aggregate the organic compounds into classes to make the numerical solution within gridded air quality models a tractable problem. One compression technique entails lumping similar organic (carbon-hydrogen) structures together as defined by the bonds between carbon atoms. A series of mechanisms evolved known as Carbon Bond (latest version is CB4). Another compression technique lumps organic compounds of similar reactivity together into certain classes. The series of mechanisms developed at the Statewide Air Pollution Research Center (SAPRC) in California uses this method (latest version is SAPRC-99) as does the chemical mechanism developed for the RADM model (latest versions are RADM2, RACM).

Wet and dry deposition models have relevancy to the determination of the fate of all chemical species emitted into and formed within the atmosphere, for without these removal processes, pollutant concentrations would ever increase over time. For local-scale modeling assessments, these removal processes are only significant for very brief periods when wet deposition occurs, or for very large particles where gravitational settling is significant. Beginning with the assessment for acid deposition of sulfates, which was completed in 1990, it became clear that improvements were needed in the roles played by cloud processes in removal of species by wet deposition. Then with the current interest in the characterization of fine particulate (aerosols) concentrations, wet and dry deposition models have become critically important for the proper partitioning of species between gas-phase and particle-phase.

Model Evaluation

The terms "evaluation", "verification", and "validation" are being used to identify the process of comparing model estimates with observations. The validity of a model is examined by assessing how well model predictions agree with observations given a perfect specification of model inputs. Since the model inputs are less than perfect, can the models be validated? The term "verification" usually applies to models that are used in the prognostic mode (e.g., a weather prediction model). The term "evaluation" entails a systematic comparison of the model predictions against observations. Two kinds of model evaluations are being typically performed: performance evaluations and diagnostic evaluations. Performance evaluations and diagnostic evaluations assess different qualities of how well a model is performing, and both are needed to establish the model's credibility within the client and scientific community. Performance evaluations allow us to decide how well the model simulates the temporal and spatial features imbedded in the observations. Performance evaluations employ large spatial/temporal scale data sets (e.g., national data sets) and allow the determination of the relative performance of a model vis-a-vis alternative models by comparing end-to-end model calculations against full-scale data sets. Diagnostic evaluations allow determination of model capability to simulate individual processes that affect the results (e.g., droplet fall velocity using small-scale data sets such as from, special field experiments, wind tunnels or other laboratory equipment). When model formulation and model inputs are precise, diagnostic evaluations allow us to decide if we get the right answer for the right reason, and usually employ smaller spatial/temporal scale date sets (e.g., field studies). Although different in analysis scale and technique, these two evaluation types will require the same basic protocol, namely, data collection and organization, analysis development and application, and the dissemination of results to both client and scientific communities.

In the past, the emphasis of the statistical evaluation comparisons has been on the "intended use." For instance, one of the uses for modeling results is to estimate the highest concentration values to be expected over a 5-year period, resulting from the operation of a proposed new power plant. Other statistical measures have also been employed to compare the concentration (or dose) values of "intended use," such as number of values within a factor of two, linear least-square fits to scatter plots of observed and predicted values, and normalized mean-squared errors of observed and predicted values. Implicit in such statistical comparisons is an assumption that the predicted and observed distributions of concentration values are from the same population, which may not be a well-founded assumption. Work is underway to develop a new generation of evaluation metrics: ones that diagnostically probe a model's chemical and thermodynamic processing and ones that take into account the statistical differences (in statistical deconvolutions and error

distributions) between model predictions and observations. As a result, a shift in philosophy is occurring as to how models of environmental processes can be acceptably evaluated. Most models provide estimates of the first moment of conditions to be expected for each ensemble (e.g., average time-space variation of the meteorological conditions, average time-space variation of the surface-level concentration values). The key to the next-generation evaluation metrics is that they will no longer assume that the modeled and observed values come from the same statistical population of values. They will assume that they "share" certain fundamental properties, but could be inherently different. A next step is to develop spatio-temporal analysis evaluation procedures that compare the modeled and observed spatial and temporal patterns of dispersing material. Concentration patterns are known to contain correlation structures in time and space that could be tested. For instance, the time series of ozone and particulate observations contains fluctuations occurring on many different time scales. Since these observations are taken at discrete time intervals, the sampling interval limits the highest frequencies that can be observed, and the sampling duration limits the lowest frequencies that can be observed. Various authors have illustrated the use of spectral decomposition of a time series of ozone concentration values to investigate model performance at selected time scales of interest. Research is being conducted to explore other methods for the statistical comparison of modeled and observed spatial and temporal behavior of pollutant concentrations.

Model Applications

To date, air quality models have been used primarily in the retrospective mode, to simulate historical episodes and to approve or deny a new source permit or an emissions management plan to meet and maintain the relevant NAAQS. From the discussion above, it is clear that models cannot be relied upon to predict the absolute concentration values at a given time for a location. Since not only the model formulations are less than perfect, the input data for the models is uncertain. Therefore, models are more reliable when used in a relative rather than in absolute sense. When the air quality models are used in the regulatory framework, it is important to consider probabilistic-based decision-making. Hence, outputs from deterministic models need to be transformed into the probabilistic form so the model results can be used more confidently in making emissions management decisions. In other words, policy makers need to see and understand the relative efficacy of various emission control strategies in meeting the NAAQS and the probability of the success of the adopted control option in maintaining the NAAQS over a region. Such integrated observational-modeling techniques are emerging now to help address the new ozone and PM standards in the United States.

Future Needs

In the future, we see several areas for air quality model research. These include on the large scales, addressing modeling issues associated with cross-boundary transport on hemispheric and global scales, and on fine scales, the role of models for driving human exposure assessment, especially in the high population urbanized areas. Research is underway to determine the limits of Eulerian grid models to provide operational estimates of fine-scale grid estimates of the concentration values. Then, the goal is to 'add' to this solution a stochastic component that represents the sub-grid variability in the predicted concentration values to enable the assessment of populations to exposures of various pollutants at the neighborhood scale. Also, attention to modeling emissions from natural and prescribed fires, from wind blown dust, and from marine sources such as sea salt are needed. Further, some classes of "emissions" should be handled within the chemical-transport model, because some compounds may be either emitted or deposited (bi-directional flux). Emissions and dry deposition are currently treated separately in air quality models, but future models should consider bi-directional fluxes of compounds such as ammonia, mercury, and trace gases in a holistic manner.

Work is underway to apply numerical models to routinely forecast air quality over the United States. To this end, ensemble modeling might be needed to improve the quality of the predictions. These efforts, in turn, would help us in developing the next generation of coupled meteorological-chemical transport models. It is important to realize that the new model evaluation metrics will require dense field data sets that are of sufficient length in time to support spectral decomposition analyses, or sufficient extent to define samples from ensembles for analysis. To this end, organizing and making publicly available field data from past model evaluation exercises is important, so that research can be conducted towards best use of these often episodic data sets, in light of the data needs of the new model evaluation procedures.

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16. ABSTRACT

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