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Meteorological modeling for air-quality assessments[☆]

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Abstract

Meteorological fields are required inputs for air-quality models, but they can contain significant errors which contribute to uncertainties in simulations of airborne chemical species, aerosols and particulate matter. Atmospheric states can be diagnosed from observations or simulated by dynamical models (with or without four-dimensional data assimilation, FDDA). In general, diagnostic models are straightforward to operate, but obtaining sufficient observations to analyze regional-scale features is costly, may omit key variables and often lack sufficient spatial or temporal density to describe the fields adequately. Dynamical models, although still imperfect, have improved in recent years and are now widely accepted for many air-quality modeling applications. Examination of the current state of dynamical models used as meteorological pre-processors indicates that useful simulations for real cases are feasible for scales at least as fine as 1 km. Introduction of faster computers and practical FDDA techniques already allow simulations of regional episodes lasting up to 5–10 d with fine resolutions (5 km or less). As technology has improved, however, a need has developed for better parameterizations to represent vital physical processes, such as boundary layer fluxes, deep convection and clouds, at these finer grid scales. Future developments in meteorological modeling for air-quality applications will include advanced model physics and data assimilation, better coupling between meterological and chemical models, and could lead eventually to widespread use of fully integrated meteorological-chemical models for simulating and predicting air quality. © 2000 Elsevier Science Ltd. All rights reserved.

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1. Introduction

Meteorology is well known to be an important factor contributing to air quality. It encompasses many atmospheric processes that control or strongly influence the evolution of emissions, chemical species, aerosols and particulate matter. These processes include horizontal and vertical transport, turbulent mixing, convection and lightning-induced generation of nitrogen oxides (NO_x), and both dry and wet deposition to the surface. In addition, the rates at which secondary species and aerosols form and certain chemical reactions take place are affected directly by the relative humidity, solar energy, temperature and the presence of liquid water (clouds). Because trace gases and particulates exist in minute concentrations, measurements are often difficult and costly to obtain, and generally are too sparse to adequately define the ambient state. Therefore, detailed Eulerian numerical air-quality models (AQMs) have been developed for scientific investigations and to support emissions-control policy decisions. They also provide one of the only methods to predict and evaluate the possible impacts of proposed emission reduction strategies. Although they do not replace other methodologies, advanced models have become valuable and widely used tools for air-quality applications.

Nevertheless, AQMs are extremely complex and their skill depends on the accuracy of a large number of internal and external parameters. Despite advances in

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computer technology, data collection and numerical modeling techniques, performance evaluations of state-of-the-science air-quality models demonstrate that their solutions can contain important errors (Russell and Dennis 2000). The causes of these errors are highly complex and are only partly understood. The most important include imperfect knowledge of the initial and boundary concentrations for airborne species, the pattern and rate of emissions, and some of the atmospheric chemical reactions themselves.

Moreover, meteorological fields supplied to air-quality models may contain significant uncertainties which adversely affect model simulations (e.g., Sistla et al. 1996). Although additional quantities are required occasionally, the principal meteorological state variables needed for AQMs are horizontal and vertical wind components, temperature, water vapor mixing ratio, cloud fraction and liquid water content, precipitation, solar actinic flux, sea-level pressure, boundary layer depth, turbulence intensity (turbulent kinetic energy or vertical diffusion coefficient), and surface fluxes for heat, moisture and momentum. The rate at which air is processed through convective clouds, including entrainment and detrainment rates, also can be very useful for many applications. Furthermore, models designed to simulate biogenic emissions are highly sensitive to the accuracy of meteorological inputs, especially the temperature in the plant canopy.

The *objectives* of this review, then, are *to document current alternatives used to define meteorological fields for air-quality assessments, to discuss their capabilities and limitations, and to identify areas for future improvements*. These topics, of course, are of such broad interest that many hundreds of scientists are actively involved in related research and applications worldwide. It is impossible to review adequately all recent work in a single paper. Thus, the scope of discussion is limited primarily to work in North America, although this gives inadequate recognition to many investigators in Europe, Asia and elsewhere, who have contributed much to our knowledge. Even in North America, it is difficult to discuss fully all of the most recent progress. Thus, this present paper seeks to summarize the more important advancements in a representative manner.

1.1. Motivation for a critical review

A review of meteorological processors used in air-quality studies is appropriate at this time from several perspectives. First, many proposed pollution-abatement strategies designed to meet mandated air-quality standards could have far reaching socio-economic consequences. The cost of implementing a particular strategy requiring, say, 50% lower emissions for volatile organic compounds (VOCs) may affect certain industries far more heavily than others. Among the many consequences could be the loss (or creation) of jobs and changes in tax structures, consumer prices, health-care costs, global and regional environmental quality, and international economic competitiveness. These legitimate and sometimes competing values may be difficult to quantify, but must be weighed carefully by policy makers. Therefore, there is a great demand for more precise information about factors contributing to air pollution and the impact of proposed controls. The guidance supplied by AQMs certainly is very valuable, but the errors found in some applications to historical cases often are a concern when current models are used for regulatory purposes. Since meteorology is a primary factor affecting both actual and simulated air chemistry, it is vital to assess current model skill and to identify new approaches likely to improve their accuracy in future studies.

In the past, health impacts from ozone were considered significant only when hourly averaged peak exposure exceeded 120 ppb. It is now believed that exposure to lower dosages of ozone and particulate matter over longer periods can be as detrimental as short-term high dosages. Consequently, the US Environmental Protection Agency (EPA) has sought to mandate a new ozone standard of 80 ppb over eight hours. Most advanced meteorological modeling systems, however, have been designed to study short-term cases on order of one to several days. The growing concern about seasonal and annual exposures necessitates a re-examination of current modeling approaches.

Moreover, the rapid growth in computer technology has led to finer-resolution models with more complex physics, numerics and four-dimensional data assimilation. As dynamical models have become more accurate, they have steadily replaced simpler diagnostic approaches. These new developments in numerical methodologies make it important to step back and re-assess model capabilities. Finally, as new remote sensing instruments have become available, vastly more meteorological data can be collected than was feasible in the past. Methods must be developed to better exploit these new data. The implications of these challenges will be explored in the sections to follow.

1.2. Background

The weather affecting a particular locale at a given time is the result of numerous processes acting on a range of scales. Atmospheric motions, for the present discussion, can be separated into two classes: *wind* and *turbulence*. Wind is the deterministic three-dimensional (3-D) motion field with time scales from minutes to days and having spatial scales from kilometers to the planetary scale. Atmospheric turbulence, on the other hand, consists of the chaotic (non-deterministic) motions resulting from instability in a fluid at high Reynolds numbers. It consists of eddy motions having scales from a few millimeters up to perhaps a kilometer, and with time scales from about a second to 20–30 min. With respect to airborne chemical species, the wind is mostly responsible for transport, while turbulence results primarily in mixing of constituents.

Many high-pollution episodes are warm-season cases with relatively weak dynamics (light winds, little precipitation, and moderate to shallow mixing depths). They tend to be associated with broad high pressure systems having weak horizontal pressure gradients (e.g., Zhang and Rao, 1998). In such weakly forced situations, turbulent motions may be of the same order as the wind speed, so wind directions are highly variable. Even modest directional errors on order of 10°, persisting over time, can result in large errors in estimations of regional-scale plume trajectories (Warner et al., 1983). Thus, simulation of high-pollution episodes with weak dynamics can be an especially challenging meteorological problem.

Within the air-quality community, the two general classes of atmospheric motions (wind and turbulence) are often divided into four scales: large, regional, local and turbulence scales. In the broader meteorological community, however, the same range of motions generally is described by six scales: global, synoptic, mesoalpha, mesobeta, mesogamma and turbulence scales (Orlanski, 1975). The relationships between the spatial and temporal ranges of these scales are given below, along with a brief description of their influence on air quality.

At the *turbulence scale* (*microscale*) in the planetary boundary layer (PBL), stack-plume characteristics are described well by the intensity and time during which vertical and horizontal mixing and eddy transport act on recent emissions, while transport by the mean wind may be considered to be quasi-steady (Pasquill and Smith, 1983). The important length scales range from about 1 mm to 2 km. It should be noted that, above the PBL, turbulence also can be used to describe the mixing within convective clouds, and meteorologists sometimes refer to scales of about 0.5–5 km as the *cloud scale* (Ray, 1986). Cumulus clouds can be important for venting pollutants from the PBL and later releasing them into the free troposphere, where they can transported over long distances.

The *local* (*mesogamma*) scale, on the other hand, involves spatial scales of 2–20 km and time periods from minutes to several hours. During that time, individual plumes often can be identified easily and associated with their specific sources. However, plume characteristics gradually become more a function of transport irregularities in the 3-D wind and the evolution of atmospheric chemistry, rather than simply due to mixing of the original emissions by turbulent eddies.

On the *regional* (*mesoalpha and mesobeta*) scale, individual plumes from specific point sources already have interacted such that they become steadily more difficult to associate with their sources, except perhaps by using source-specific tracers (e.g., heavy metals from an ore smelter). The mesobeta scale covers 20–200 km, while the mesoalpha scale encompasses 200–2000 km. Regional transport times range from several hours to a couple of days. Eulerian air-quality models for the local and regional scales generally assume plumes have already dispersed out to the resolved grid scale (several kilometers) or else use a plume-in-grid sub-model to represent concentrated point sources.

Finally, the influence of scales larger than the mesoscales cannot be ignored. The *large* (*global and synoptic*) *scale* is important for determining the chemistry "background" state often used to describe lateral boundary conditions in regional studies. For example, the chemical background may be defined in terms of mean concentrations expected upwind of a continent, where it is assumed that a quasi-equilibrium has been established. The synoptic scale covers 2000–6000 km and time scales of 1–4 d. The global scale extends over 6000–40000 km and time periods of at least 2–10 d.

Some additional implications of scale-dependent meteorological processes for air-quality modeling were discussed recently by Pielke and Uliasz (1998). Over the past two decades, most major air-quality field studies [e.g., Lake Michigan Ozone Study (LMOS), SJVAQS/AUSPEX[1] Regional Modeling Adaptation Program (SARMAP), North American Research Strategies for Tropospheric Ozone – Northeast (NARSTO-NE), South Coast Air Quality Study – 1987 (SCAQS-87), and Southern California Ozone Study – 1997 (SCOS97)], and numerical modeling applications based on these programs, have been conducted at the mesoscales.

2. Methodologies for generating meteorological fields

Meteorological processors used to supply fields to Eulerian air-quality models can be grouped into three types. First, *diagnostic* (*or kinematic*) models are those which analyze observations taken at discrete points in time and space. Second, *dynamical models* are those which integrate the non-linear hydrodynamic equations of motion or their derivatives in a numerical framework. The third approach, utilizing *four-dimensional data assimilation*, is intended to combine the best features of diagnostic and dynamical approaches by integrating a numerical model in which data are included throughout the integration period. (Although the data-assimilating numerical models are actually a variation of dynamical models used in "hind-casting", they are considered separately here because of their emerging importance for air-quality applications.) All three methods produce gridded

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[1] San Joaquin Valley Air Quality Study/Atmospheric Utilities Signatures, Predictions and Experiments.

fields representing the key variables required by the air-quality models.

2.1. Diagnostic models

A key requirement for diagnostic models used in air-quality studies is that they provide *dynamically consistent fields* of the analyzed meteorological variables. Failure to ensure intervariable consistency in the meteorology can lead to errors in the chemical mass budgets of an associated air-quality model. Diagnostic models can be built around either univariate or multi-variate objective analysis schemes. Multi-variate schemes apply relationships between variables based on a simplified form of the hydrodynamic equations, while univariate schemes do not. Perhaps the most common constraint for air-quality applications uses the continuity equation to diagnose vertical winds from the divergence of the horizontal wind field. Such a "kinematic" approach is generally necessary because vertical velocities are mostly unobserved. Unfortunately, the divergence itself is small compared to the wind, so that local effects and wind measurement errors often give apparent divergences that exceed the true mesoscale and large-scale divergences that are related to the scales of vertical motions to be diagnosed. A multi-variate analysis may seek to limit these problems by requiring that the wind field be in balance with the mass field (e.g., geostrophic or gradient wind balance). These balance constraints, however, are not appropriate for local or urban scales (smaller than the Rossby radius of deformation). In addition, because the balance in diagnostic models does not include all terms of the dynamical equations, the resultant fields are unlikely to be dynamically consistent with the atmosphere (Moran et al., 1991).

Diagnostic models can be designed to include the effects of topography. For example, the California Institute of Technology (CIT) diagnostic wind model developed by Goodin et al. (1979,1980), produces mass-consistent urban-scale wind fields with variable vertical resolution. Following adjustment of the horizontal winds for local terrain barriers, anomalous horizontal divergences are removed through the continuity equation by adjusting the diagnosed vertical velocities. Mass is conserved on the domain, accounting for flow through the boundaries, while not imposing mass conservation locally. This approach, however, can lead to unrealistic "residual" vertical velocities at the top of the diagnostic model domain.

A more complete and widely used diagnostic model is CALMET (Scire et al., 1997a; Scire and Robe, 1997). CALMET has continued to evolve and now includes complex terrain, slope-flow algorithms for the interaction of solar geometry with terrain orientation, boundary-layer modules for land and sea, and a background ("first-guess field") that can be based on winds fields from a dynamical model. It also can be applied with a dispersion model, CALPUFF (Scire et al., 1997b). Another diagnostic system designed for calculating fine-scale transport in complex terrain, ATMOS1, was developed by Davis et al. (1984). This model uses a 3-D variational analysis technique to analyze the wind field (Section 2.3) with minimization of divergence as an analysis constraint. When coupled with a diffusion parameterization (Section 4.3), this system is used to simulate plume dispersion (King and Bunker, 1984). A recent review of diagnostic models and analysis techniques was provided by Ratto et al. (1994).

Diagnostic models are inexpensive to operate and generally require little specialized training. They do not involve time consuming integrations of nonlinear equations, so they are attractive for use in real-time emergency rapid-response plans. Another advantage is that, assuming sufficient observations exist to perform an analysis, all of the data can be utilized. Finally, because each analysis is generated with a fresh set of observations, there is no accumulation of errors at successive times through an episode.

Diagnostic models, however, have several disadvantages. First, they generally are based on an incomplete or idealized set of equations. Thus, whatever balance is imposed between variables does not reflect all of the forces or processes acting in the environment and, hence, realistic intervariable consistency often cannot be attained. Some variables important for AQMs, such as clouds and vertical velocities, may be so poorly observed that direct analysis is impractical. Additionally, diagnostic models often have difficulty representing flows accurately in data-sparse regions (e.g., mountains or oceans) and their analyses may have inappropriately smooth structure.

Their most important limitation, however, is that they cannot have greater detail than that resolvable, in space and time, by the observation set. Routine observations from the National Weather Service are insufficient to resolve local-scale features and many regional-scale features, as well. This is especially true above the surface. Therefore, major research programs often need special observing systems to enhance the data density and frequency. However, even when limited to fairly brief intensive observation periods (IOPs), special measurement programs can cost millions of dollars and may still fail to provide all of the data necessary to describe mesoscale structures accurately. Many of these 3-D features, such as sea breezes and low-level jets, have scales requiring almost hourly measurements and resolutions of 50 km or less to define their structure and evolution. Only a few regional field studies (e.g., SJVAQS/AUSPEX in the San Joaquin Valley and LMOS in the upper Midwest) have had upper-level networks approaching this resolution (Ranzieri and Thuillier, 1991; Bowne and Shearer, 1991).

Lastly, diagnostic models have limited flexibility for sensitivity evaluations. That is, they can represent only the original set of processes contained in the data. It is generally impossible, using this method, to isolate the effects of individual meteorological processes to study their impact on air chemistry. Despite their limitations, however, diagnostic models remain valuable analysis tools useful for at least some air quality applications (e.g., when annual or interannual calculations are needed at fine scales).

2.2. Dynamical models

Dynamical models usually are based on the complete set of primitive equations for hydrodynamic flow, scaled for atmospheric applications and written in finite-differenced form. Occasionally, a non-primitive framework, such as the vorticity-mode model of Schayes et al. (1996), has been used successfully in air-quality research. While the original differential equations of these models generally are conservative for mass and energy, exact conservation may be lost when they are written as finite differences. Many of the dynamical meteorological models widely used in air-quality studies were designed originally for weather forecasting or the study of severe weather. Their development history usually reflects a dominant focus on problems related to strong dynamical forcing and deep convection. Adaptation to provide input for air-quality models is fairly simple, but requires very good physical parameterizations to ensure accurate solutions for cases with comparatively weak dynamics (Section 4.3). However, small mass-budget errors may exist, especially if the equations are not written in mass-flux form (Byun, 1999a,b).

Limited-area applications of dynamical models represent complex initial-boundary value problems in nonlinear partial differential equations. First, data are analyzed to supply the model with gridded observed fields (minimally, the wind, temperature, water vapor and pressure) at the selected initial time. For typical air-quality studies, cases are run well after the actual events occur, so similar analyses can be used to supply observed lateral boundary conditions at prescribed intervals (perhaps 12 h or less). These analyses are then interpolated to provide lateral boundary conditions at each time step. It is the unavailability of observed lateral boundary conditions, of course, that distinguishes a true *forecast* from an atmospheric *simulation* (which can be a forecast or "hindcast").

Numerical model frameworks can be either *hydrostatic* or *non-hydrostatic*. In hydrostatic models only gravity and vertical pressure gradient forces are retained in the third (vertical) equation of motion. This assumption simplifies the primitive equation set and is appropriate for scales greater than about 10 km (Pielke, 1984). Examples of hydrostatic dynamical models used for air-quality studies include the National Centers for Atmospheric Prediction's (NCEP) operational Eta model (Mesinger et al., 1988; Black, 1994) and the model of Lu and Turco (1995), plus the research-based Penn State University/National Center for Atmospheric Research (PSU/NCAR) mesoscale model, MM4 (Anthes et al., 1987) and the Colorado State University Mesoscale Model, CSU-MM (Mahrer and Pielke, 1977; McNider and Pielke, 1981). The latter two older models have been "frozen" and no longer undergo active development at their parent institutions.

At smaller scales, vertical accelerations cannot be ignored compared to gravitational and pressure gradient forces, and the equation set is said to be non-hydrostatic. As computer resources expand and demand grows for finer-scale numerical products, non-hydrostatic models have become the dominant framework used in dynamical models. These models usually have a nested-grid capability, terrain-following vertical coordinates, flexible resolution and a variety of physical parameterization options. The research-grade non-hydrostatic models most commonly used in the US for air-quality applications are the PSU/NCAR Fifth Generation Mesoscale Model (MM5, Grell et al., 1994) and the Colorado State University Regional Atmospheric Modeling System (CSU-RAMS, Pielke et al., 1992; Nicholls et al., 1995). In Europe, the EURAD air-quality modeling system uses a variant of MM5 as its meteorological driver in a coupled nested-grid numerical framework (Jakobs et al., 1995).

Three recently released non-hydrostatic models also are readily adaptable for air-quality studies. In the US these are the University of Oklahoma's Atmospheric Regional Prediction System (ARPS, Xue et al., 1995) and the Navy's Coupled Ocean-Atmosphere Mesoscale Atmospheric Prediction System (COAMPS, Hodur, 1997). For example, COAMPS has been applied to study toxic-plume trajectories that could have been associated with the Gulf War illnesses in Kuwait (Westphal et al., 1999). In Canada, the Mesoscale Compressible Community model (MC2) has been developed for general fine-scale meteorological applications (Benoit et al., 1997) and recently has been used to provide meteorological fields for an AQM (Mailhot et al., 1998). Table 1 summarizes the characteristics of these five non-hydrostatic models, each of which continues to undergo development. Although all five models have broadly similar characteristics, the MM5 and RAMS currently are the most thoroughly tested models for air-quality studies. Finally, the US EPA has incorporated the MM5 into its framework modeling system, called Models-3, which is designed to be a plug-compatible platform for testing and comparing emissions models, meteorological models and chemistry models for air-quality applications (Byun et al., 1998). EPA intends to incorporate several additional model options, including the RAMS meteorological driver, into the Models-3 framework.

Table 1
Characteristics of non-hydrostatic modeling systems suitable for air quality applications

Model name	Basic equations	Horizontal grid	MAP projection	Vertical coordinate	Surface scheme
ARPS (U.Okla)	Compressible, time splitting	Arakawa-C	Lambert con., Polar stereo., Mercator	Terrain following height	Prognostic surface temperature and moisture
COAMPS (US Navy)	Compressible, time splitting	Arakawa-C	Lambert con., Polar stero., Mercator, Spherical	Terrain following height	prognostic surface temperature and moisture
MC2 (Environ. Canada	Compressible, time splitting semi-Lagrangian	Arakawa-C	Polar stereo., Mercator	Terrain following height	Prognostic surface temperature
MM5 (PSU/NCAR)	Compressible, time splitting	Arakawa-B	Lambert con., Polar stero., Mercator	Terrain following pressure	Prognostic surface temperature, NCEP/Oregon State Univ. soil moisture/vegetation
RAMS (CSU)	Compressible, time splitting	Arakawa-C	Rotated polar stereographic	Terrain following height	Prognostic surface temperature, and moisture

Model name	Boundry layer physics	Explicit moist physics	Deep convection	Atmospheric radiation
ARPS (U.Okla)	1.5-order TKE	Liquid, ice, & mixed phase	Kain-Fritsch, Kuo	Column long- and short-wave with cloud effects
COAMPS (US Navy)	1.5-order TKE	Liquid, ice, & mixed phase	Kain-Fritsch	Column long- and short-wave with cloud effects
MM5 (PSU/NCAR)	1.5-order TKE, Blackadar non-local	Liquid, ice, & mixed phase	Kain-Fritsch, Grell, Betts-Miller, Anthes-Kuo, Modified Arakawa-Schubert, Fritsch-Chappel	Column long- and short-wave with cloud effects
MC2 (Environ. Canada)	1.5-order TKE	Liquid, ice, & mixed phase	Kuo, Kain-Fritsch, Fritsch-Chappell	Column long- and short-wave with cloud effects
RAMS (CSU)	1.5-order TKE	Liquid, ice, & mixed phase	Modified Kuo	column long- and short-wave with cloud effects

All of these meteorological research models have been adapted to generate routine real-time forecasts (semi-operationally) at various institutions. Recently, several research groups in the US and Europe have begun work to couple meteorological and chemistry models to produce real-time numerical air-quality forecasts (McHenry et al., 1999). Although initially intended for research purposes, the potential value of real-time air-quality forecasting could be enormous. Continuous application and evaluation of the research models on fixed domains should lead to more robust models with lower biases.

Dynamical models have a number of advantages compared to diagnostic models. Most important, their potential for resolving regional and local-scale atmospheric circulations (at least down to scales of about 1 km) is limited only by the availability of computational resources. They are more costly to operate than diagnostic models, however, because their solutions require integration of non-linear equations over many small time steps. While this handicap will never disappear entirely, it is becoming much less severe as the price for computers rapidly falls and their performance soars. Already, modestly priced desktop personal computers with multi-processor capabilities are sufficiently advanced to integrate dynamical models at speeds possible only on mainframe supercomputers until about 1995.

Another advantage is that they do not require such an extensive (and expensive) observation network to obtain products with the same resolution as a diagnostic model. Given appropriate synoptic-scale initial conditions, dynamic models with finer grid resolutions actually generate regional and local-scale features unresolved in the data. Development of fine-scale structure is due both to the model's resolved topographical forcing and internal dynamical forcing (Anthes, 1983). Also, the influence of sub-grid scale physical processes can be represented in these models through parameterizations. Important processes requiring parameterizations in regional and local scale models include deep moist convection, fog, precipitation microphysics, shallow clouds, radiative processes, surface fluxes and turbulence (Section 4.3). Furthermore, dynamical models can serve as virtual atmospheric laboratories in which repeatable experiments can be performed to isolate the role of individual physical processes and, in conjunction with AQMs, to evaluate the sensitivity of air chemistry to different forcing mechanisms.

The most important disadvantage of traditional dynamical models (without four-dimensional data assimilation) is that, other than at the initial time, observations are ignored (except for validation purposes). Consequently, imperfections in the model's numerics, physics, or initial conditions cause errors that can accumulate over time. The application of observed lateral boundary conditions causes internal errors to be swept out of the domain in long hindcast integrations by the large-scale mean wind field, while more accurate conditions are swept in at the upstream boundaries. Nevertheless, the error accumulation in regional and local-scale domains can be severe enough to make the solutions of decreasing practical value for air-quality applications after about 48 h (Stauffer and Seaman, 1990; Seaman et al., 1995). For air-pollution episodes of 4–6 d, this represents one of the most severe problems encountered in air-quality studies. Some typical skill scores for dynamical models are given in Section 3.

Finally, the dynamic meteorological models are highly complex numerical systems requiring fairly extensive training to operate and to interpret accurately. Trouble shooting in codes of perhaps 50,000 lines or more can be a daunting task, even with the aid of documentation and user's manuals. Adaptation of a modeling system to a new region, perhaps with nested domains and at different resolutions, requires in-depth understanding of the limits of parameterization applicability. Also, real-case applications involve strict observation quality-checking requirements, since erroneous data can trigger amplifying model errors. Nevertheless, despite these difficulties, dynamical models in the context described here have enjoyed some success when used for air-quality applications.

2.3. Data assimilating models

Four-dimensional data assimilation (FDDA) is a method in which the growth of errors in a dynamical model is limited by allowing observations distributed in space and time to correct for errors in the model's solutions. Data assimilation has become common for meterological model applications in recent years, especially in support of air-quality studies. It is a valuable technique when high-precision atmospheric fields are required, but when diagnostic and dynamical models alone are insufficient to resolve all the important features in those fields. FDDA can be applied in both hydrostatic and non-hydrostatic meteorological models. Some typical results appear in Section 3.

Data assimilating models (DAMs) have all the advantages noted in Section 2.2 for dynamical models, but they also can use many of the data throughout the integration period, rather than at the initial time only. Most importantly, FDDA reduces the accumulation of errors found in dynamical models, which can be particularly valuable for simulations longer than 48 h. For any FDDA system, care must be taken in developing the data insertion strategy that controls when and where the observations are assimilated or how strongly they affect the solutions. To be most effective the weighting strategy should allow the data's influence to be physically consistent with the dominant scales encountered in the atmosphere. Thus, an ideal radius of influence could be calculated from the error covariances, as in optimal interpolation (e.g., Daley, 1991).

Two major types of continuous FDDA are in use today: *Newtonian relaxation* and *variational analysis*. They are said to be continuous because the data can affect the solution each time step, thereby minimizing "shock" that occurs in other intermittent FDDA approaches. Newtonian relaxation, or *nudging*, relaxes the model state toward the observed state by adding an artificial tendency term to one or more of the prognostic equations, based on the difference between the two states. It is applied most often to wind, temperature and water vapor, but in principle, can be applied for any prognostic variable. However, it is not used directly for influencing diagnosed variables.

Nudging was first proposed by Anthes (1974) and was developed for air-quality modeling by Stauffer and Seaman (1990) and Stauffer et al. (1991). It can be applied either by nudging toward gridded analyses, which are interpolated to the model's current time step, or by nudging directly toward individual observations within a time-and-space "window" surrounding the data. These two approaches are referred to as "analysis nudging" and "obs nudging", respectively. Analysis nudging is ideal for assimilating synoptic data that cover most or all of the model domain at discrete intervals. Obs nudging does not require gridded analyses and is better suited for assimilating high-frequency asynoptic data that may be distributed irregularly in space and time.

The coefficient, G, which controls the magnitude of the nudging term, usually is chosen in the range $1\text{–}5\times10^{-4}\,\text{s}^{-1}$. This constrains the magnitude of the nudging term to be small compared to the major physical terms in the dynamical equations (Ardao-Berdejo and Stauffer, 1996). For brief periods, it may become large compared to the physical terms if the model's solution develops significant local errors, but in general the nudged solution remains close to the physically balanced state and retains internal consistency among the variables. The nudging approach (especially analysis nudging) is quite simple to implement in existing dynamical models. However, the development of physically realistic data-weighting strategies requires some care to prevent unrealistic local forcing or excessive smoothing. New users will benefit from a review of basic papers describing assimilation strategies (e.g., Stauffer and Seaman, 1990,1994; Seaman et al., 1995; Shafran et al., 2000).

As implied above, an important issue for any FDDA technique is whether the data are used to the greatest advantage possible. This concept of a "best fit" to the data is often expressed in mathematical terms as the minimization of the sum of the squares of errors between the model solutions and observations, distributed in space and time. That is, in the "least-squared-error" sense, optimization is attained when the model error, or the "distance" between the trajectory of the model solution (not to be confused with a particle trajectory) and the observations *throughout some finite assimilation period*, is a minimum. The DAM solution obtained from nudging usually cannot be considered mathematically optimized in this least-squares sense because no such constraint is applied during the integration. However, Lorenc (1986) has shown that the nudging approach can be made to converge toward that state. Thus, nudging has been widely tested and found to be successful in a variety of cases. When appropriately scaled for physical modes of the data, it can be used effectively for many air-quality applications.

The variational method, on the other hand, is based on optimal control theory and the calculus of variations (Sasaki, 1958). The means of evaluating the error in a four-dimensional variational solution is often through an equation called the *"cost function". If the "model" itself can be neglected as a source of error during the assimilation period* (a "perfect-model" assumption), then 4-D variational analysis determines an optimal state (i.e., analysis) for which the model errors throughout the assimilation period are minimized through an iterative process (Lewis and Derber, 1985). In addition to assimilating the primitive variables, variational analysis also can assimilate any type of non-primitive variable, as long as it can be expressed as a constraint in the cost function (Caplan et al., 1997; Derber and Wu, 1998). For example, satellite radiances can be assimilated directly instead of performing an uncertain conversion to temperatures.

The form of the cost function defining the relative weight given to each of the assimilated variables can be important to the physical consistency of the results. Zupanski (1996) has shown that relaxing the perfect-model assumption can have large effects on the resultant "optimal" analysis. This sensitivity to the balance specified between the constraints in the cost function, despite the fact that all the variational solutions are optimal in a mathematical sense, is rather similar to the sensitivity of nudging solutions to different data-weighting strategies. That is, depending on how the variational constraints are designed, any number of "optimal" states are possible. Furthermore, the perfect-model assumption is clearly invalid. It may be acceptable when used for relatively brief model integrations, but for air-quality episodes of 5–6 d in length, the perfect-model assumption could lead to serious problems (Stauffer, 1995).

Four-dimensional variational analysis can require extensive computational resources. Its efficiency can be improved somewhat by introducing an *adjoint* form of the dynamical model that can be integrated backward in time (Lewis and Derber, 1985). The adjoint model often is derived from a tangent linear model (TLM) to the forward model (e.g., Errico and Vukicevic, 1992). The TLM is a linearized version of the forward model which predicts how "small" perturbations in the model state (on order of one percent or less) will grow in time. The purpose of the backward (adjoint) model integration is to compute the gradient of the cost function (the gradient of the model output with respect to the control variable, or model input), thereby defining the "direction" in which the initial state must be changed. Calculation of an optimal four-dimensional analysis with the adjoint method may require a series of 10–40 (or more) forward/backward model integrations for well-conditioned problems. Thus, despite the acceleration of the solution by the adjoint method, a major disadvantage of 4-D variational analysis is that it remains computationally intensive compared to other methods.

There are other obstacles to using the adjoint approach for air-quality applications. Its accuracy depends in part on the following: (1) availability of a "perfect model", (2) application within the predictability limits of the modeling system (especially the TLM), and (3) difficulties in treating discontinuous "on/off" processes such as convective latent heating (Stauffer, 1995). If the model solution for a given case is ill-conditioned in any of these respects, the cost function may fail to converge to a minimum, or the solution may be unrepresentative of the actual physical state of the atmosphere.

It appears that the variational analysis technique will have to undergo several more years of development and testing before it can be used routinely for air-quality modeling studies. However, many of the present uncertainties associated with the adjoint method eventually will be overcome or mitigated. The potential of being able to produce meteorological fields which objectively provide the best possible fit to the available data will remain an important incentive to stimulate additional research.

In summary, while nudging FDDA usually is not optimal in the mathematical sense, it is comparatively inexpensive and reliable when the weighting strategy is designed to allow the model's physical terms to dominate the solutions. It has been used widely in recent years with considerable success, and will probably continue to be a valuable tool, at least until the problems associated with 4-D variational analysis can be controlled.

3. Examples and intercomparison of methodologies

A definitive comparison of skill among diagnostic, dynamical and data-assimilating meteorological models is demanding. For example, much uncertainty remains about the quality of winds from diagnostic models, because most cases in the literature use all of the available observations to generate the analyses. Without an independent data set for evaluation, it is difficult to determine their potential for error in realistic applications. In many cases, evaluation is performed by using diagnosed winds to drive a local plume dispersion model, whose results then can be evaluated against tracer observations (e.g., King and Bunker, 1984). While valuable, interpretation of this approach is complicated by the addition of the dispersion model, which may introduce errors of its own.

Another approach is to use an idealized analytic field to represent a perfectly known atmospheric state (e.g., Goodin et al., 1979). Pseudo-observations of wind can be selected from analytically generated fields to provide input to the diagnostic model. Validation is done by comparing the diagnosed winds to the original analytic field. However, idealized fields are rarely as complex as those encountered in real atmospheric cases, where nonlinear interactions, local topography and multiple physical processes become important. It remains problematic to obtain an independent data set for real-case validation of diagnostic models. The lack of adequate evaluation for diagnostic models on a case-by-case basis also was noted by Pielke et al. (1991).

Statistical comparisons between dynamical and data-assimilating models are more common and a summary from several recent studies appears in Table 2. These studies indicate that FDDA can reduce errors by about 25–60% at the regional scale (Stauffer and Seaman, 1990; Stauffer et al., 1991) and the local scale (Stauffer and Seaman, 1994; Fast, 1995). In those studies, however, the assimilated data also were used as the verification data set. Recently, Seaman et al. (1995), Michelson and Seaman (2000) and Tanrikulu et al. (2000) performed experiments in which half of the data from a special field study were withheld from the DAM and were used in an independent validation to compare model solutions with and without FDDA. These tests verified that FDDA does indeed reduce errors significantly, compared to the dynamical model approach, including the regions between sites where the assimilated data originated. Furthermore, assimilation of one-half of the observations in these data-rich cases produced about two-thirds of the improvement obtained when all of the data were assimilated.

Fast (1995) performed perhaps the only intercomparison among a diagnostic model (ATMOS1), a dynamical model (CSU RAMS) and a DAM (CSU RAMS with nudging FDDA). Using very fine horizontal resolution (1.32 and 0.33 km) to study nocturnal drainage along Colorado's Front Range during the Atmospheric Studies in Complex Terrain (ASCOT), the DAM clearly produced more accurate wind field statistics than the dynamical model alone. No similar validation was performed against observations, however, for the diagnostic wind model. Next, wind fields from ATMOS1 and the DAM were used to drive independent dispersion models and results were compared to tracer observations. Dispersion calculations based on DAM winds were based on a Lagrangian particle dispersion model (LPDM) (McNider et al., 1988). The diagnostic model's winds, on the other hand, were applied in a separate dispersion model (ATMOS2) (King and Bunker, 1984). Results of the dispersion calculations were somewhat ambiguous. The *position* of the concentration maxima were simulated better by the DAM, however, while the concentration *magnitudes* were captured better using the diagnostic model's winds. Fast (1995) concluded that the diffusion errors associated with the LPDM were more serious than the advection errors produced by the DAM with FDDA. This intercomparison is certainly very helpful, but it still lacked an independent set of wind data to evaluate the performance of the diagnostic wind model.

In a similar approach, comparisons of AQM performance have been made using different meteorological inputs from diagnostic models, dynamical models, or DAMs. Sistla et al. (1996) found that spatially varying mixing-depth fields gave more accurate ozone simulations in an AQM than did spatially invariant mixing depths. Fernau and Pai (1998) recently performed comparisons using three AQMs, two DAMs and a diagnostic model. Their results showed that by most (but not all) measures, the skill of the AQMs was improved by using meteorological fields from the DAMs. Although this type of comparison can be affected by compensating errors in the AQMs, it does demonstrate the value of the DAMs for applications of interest to the air-quality community.

The available evidence on these three types of models, and the assessment of their inherent strengths and limitations discussed in Sections 2.1–2.3, make a strong case that the DAMs are likely to outperform the diagnostic models for most general atmospheric applications unless an extraordinary data base is available. This conclusion must remain somewhat tentative until better controlled intercomparisons are performed. Nevertheless, the versatility of DAMs for investigating process sensitivity, their ability to maintain intervariable consistency, and their ability to represent vertical velocities, divergences and a variety of physical processes (convection, clouds,

Table 2
Statistical summary of meteorological model skill in selected air-quality studies[a]

Authors	Case/Location (Length)	Model/Resol.	Results	
			Dynamical model	Data assimilating model
Ulrickson and Mass (1990a,b)	15 Sept. 1981, 8 Aug 1984 LA Basin, CA (24 h)	CSU-MM (dx = 5 km, 22 layers)	Wind RMS = 1–3 m s $^{-1}$ during day, = 1.5 m s^{-1} at night. I = ∼0.7 d, ∼0.5 night. Wind direction ME = 0–10 d e.g. in day, ∼30 deg. at night	
Seaman et al. (1995)	SARMAP, 2 cases summer 1990, San Joaquin Valley, CA (120 h)	PSU/NCAR MM5 (dx = 4 km, 30 layers)	$I = 0.51$ m s^{-1}	Multi-scale nudging FDDA. Vector Wind RMS = 2.8 m s^{-1}, $I = 0.76$, Wind Direct. ME = 4.2 deg.
Lyons et al. (1995a,b)	LMOS, 4 summer cases, 1991, Lake Mich. Region (120 h)	CSU-RAMS (dx = 4 km, 31 layers)		Analysis nudging FDDA. Wind RMS = 1.69 m s^{-1} $I = 0.59$ Wind direct, ME = −13 deg.
Fast (1995)	ASCOT, 3–7 Feb. 1991, Rocky Flats, CO. (26 h)	CSU-RAMS (32 layers) (dx-1.32 km, dx = 0.33 km)	Vector Wind RMS = 3.63 m s^{-1} Vector Wind RMS = 2.58 m s^{-1}	Analysis nudging FDDA. Wind only. Vector Wind RMS = 0.93 m s^{-1}, Vector Wind RMS = 2.23 m s^{-1}
Seaman et al. (1997)	SCAQS, 26–29 Aug. 1987, LA Basin (120 h)	PSU/NCAR MM5 (dx = 4 km, 32 layers)		Analysis-nudging FDDA. Vector Wind RMS = 1.9 m s^{-1}, $I = 0.55$, Wind direction ME = ∼10 deg. in day, ∼30–90 deg. at night with very light winds.
Shafram et al. (2000)	LMOS, 2 summer cases, 1991, Lake Mich. Region (120 h)	PSU/NCAR MM5 (dx = 4 km, 32 layers)	Wind RMS = 2.63 m s^{-1} $I = 0.54$ Wind direction ME = −8.4 deg.	Multi-scale nudging FDDA. Wind RMS = 1.72 m s^{-1} $I = 0.62$, Wind direction ME = −9.2 deg.

[a] *Note*: RMS is root mean square error, I is Index of Agreement, ME is mean error.

eddies, etc.) generally make them far more attractive than diagnostic models for use in air-quality assessments. These factors lead to the fairly certain conclusion that dynamical models with FDDA (DAMs), are now the best alternative for developing accurate meteorological fields for air-quality models.

A cautionary note on two points is appropriate. First, while the statistics in Table 2 give a useful summary of some carefully validated studies involving dynamical models and DAMs, individual cases and regional topography can vary greatly in complexity. Therefore, although the table provides a reasonably objective standard of performance, statistical skill should not be considered the only criterion for success. It is always vital to reproduce case-specific mesoscale circulations and other physical structures, which may be masked by domain-averaged statistics. Finally, *it should never be assumed that FDDA can always overcome grievous errors resulting from poor-quality initial conditions, physics or dynamics in the host model to produce superior solutions. Neither is it true that any FDDA strategy will be as successful as another.* The best approach when using a DAM is always to use dynamical models which have advanced capabilities and are well tested, and then to add well-designed data-assimilation strategies. To briefly illustrate the performance characteristics of dynamical models and DAMs in complex flows, several figures are included from some recent papers. Of course, these few figures cannot show all of the progress made by numerous investigators working on a host of problems. However, they provide representative examples of the kind of detailed meteorological structures which can be captured by these models. With insufficient observations to resolve local-scale 3-D circulations, diagnostic models are unlikely to define such features adequately. Moreover, statistical summaries alone do not reveal the obvious importance of these features to air quality for the individual episodes.

First, Fig. 1 compares two sets of model-generated backward trajectories calculated over a 36-h period during winter using a 10-km version of the PSU/NCAR MM5 (Stauffer and Seaman, 1994). This study was related to visibility-degradation research in the Four Corners-Grand Canyon region of the Southwest US. Fig. 1a (without FDDA) shows that three parcels arriving at 500 m AGL over the Grand Canyon all had sources far to the east over the Great Plains, while Fig. 1b (with FDDA) indicates shorter trajectories (weaker winds) and widely dispersed source regions inside the domain. Observations reveal a diffuse mesoscale low with three troughs and weak cyclonic low-level flow were passing slowly over the area during the study period (not shown). The use of FDDA in this case corrected the path of the low in the numerical solutions, which resulted in a reduction in vector wind errors of nearly 50%, compared to the run without FDDA.

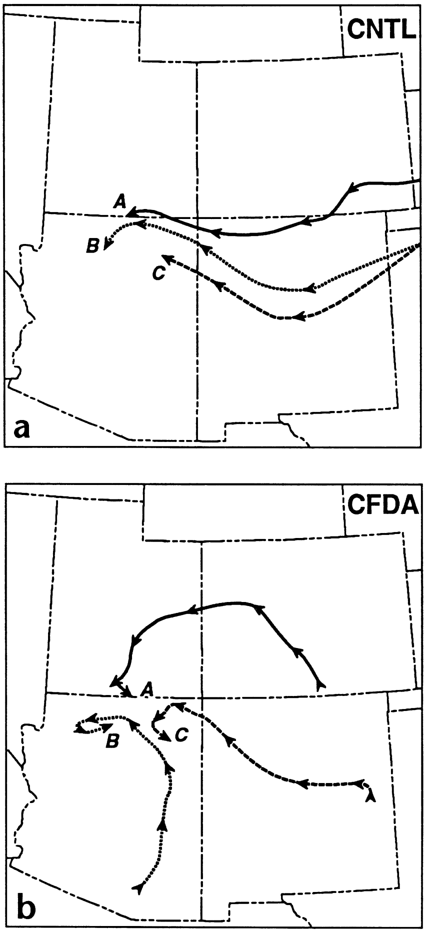
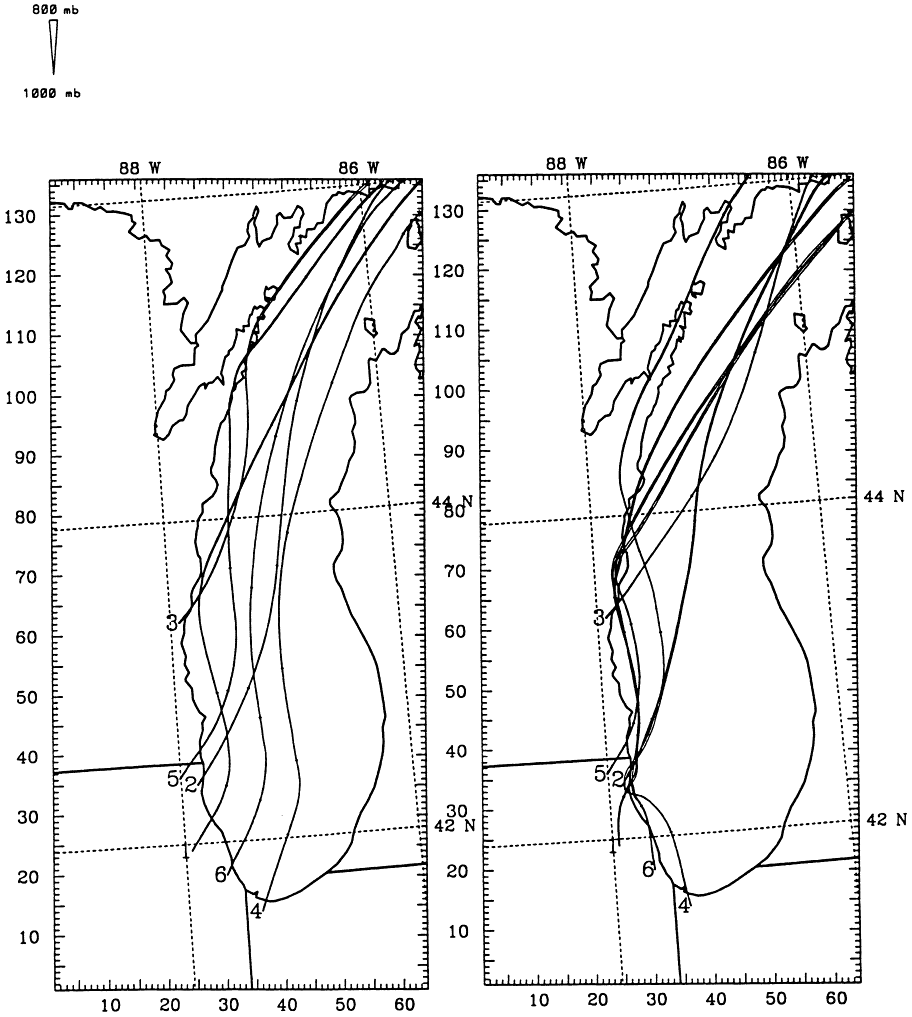


Fig. 1. MM5 backward trajectories of air parcels arriving 500 m above the surface after 36 h (at 1200 UTC, 19 January 1990) at Page, AZ (Point A), Hopi Point, AZ (Point B) and Black Mesa, AZ (Point C). Arrowheads indicate parcel direction and 6-h displacements. (a) No FDDA, (b) with FDDA (combination of analysis nudging and observation nudging) (from Stauffer and Seaman, 1994).

An example of model sensitivity to local surface temperature and moisture fluxes, as well as FDDA, is shown in Fig. 2 for a lake-breeze case on 16 July 1991 from the LMOS study (Shafran et al., 2000). In this high-ozone episode with weak southwesterly synoptic flow, the MM5 model was run at 4-km resolution both with and without nudging FDDA. Both experiments exhibited a well-developed lake breeze along the western shore of Lake Michigan in response to the surface fluxes (not shown). Correction of minor errors in the regional-scale wind directions by the FDDA, however, led to a significant change of the lake breeze flow, despite the near absence of assimilated data over the lake itself. Fig. 2 shows the impact of FDDA on 3-D forward trajectories



CNTL

GSMSFDA

Fig. 2. MM5 forward 3-D trajectories calculated for parcels released from 100 m AGL at 1200 UTC, 16 July 1991, based on wind fields from a 4-km domain. Initial release points are numbered: 1 is O'Hare Airport, 2 is Zion, IL, 3 is Milwaukee, WS, 4 is Gary, IN, 5 is inland from Zion, IL, and 6 is downtown Chicago, IL. (a) No FDDA, (b) multi-scale FDDA with combined analysis nudging and observation nudging (from Shafran et al., 2000).

in the two experiments on this day, when 1-h peak ozone exceedances were measured at numerous sites along the shore all the way to the northern Door Peninsula (not shown). For the six parcels released at 100 m AGL from sites between Gary, IN, and Milwaukee, WS, the trajectories show that the FDDA-assisted winds cause the urban plumes to become trapped in the lake breeze, where they can be re-circulated at least twice as the parcels are carried northward, while the non-FDDA winds allow emissions from many of the urban areas to travel northward up the center of the lake without being recirculated over the western shore. This recirculation wind pattern occurs in the model as a result of both the mesoscale physics and the FDDA, even though there are insufficient observations to diagnose such a complex flow directly from the data.

When a dynamical model or DAM is coupled to a dispersion model, realistic plume dispersion patterns can be calculated. Fig. 3 shows three examples of urban and utility-plant plumes simulated in the southern Appalachian Mountains by Mueller et al. (1996) using a 4.5-km version of the CSU-RAMS model with analysis-nudging FDDA and a Lagrangian particle dispersion model, or LPDM (McNider et al., 1988). This approach reveals plume trajectories and interactions as they would appear within an AQM for a non-reactive material.

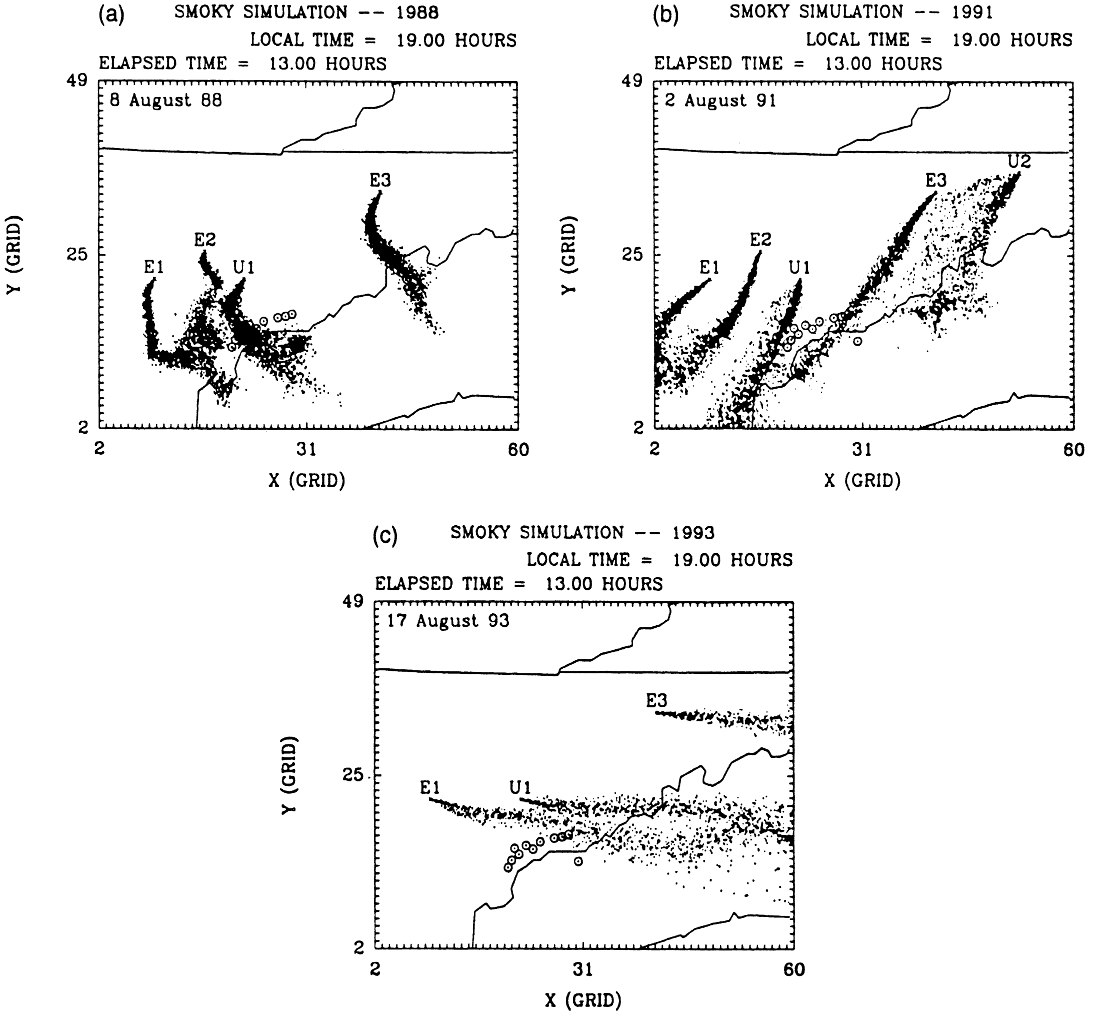


Fig. 3. RAMS-LPDM tracer-plume plots simulated for major NO_x sources in the southern Appalachian Mts. during three high-ozone episodes. (a) 8 August 1988, (b) 2 August 1991, (c) 17 August 1993. Plumes are initiated at 0600 EST (1100 UTC) and are terminated at 1900 EST (0000 UTC) (from Mueller et al., 1996).

Next, Fig. 4 shows an isentropic cross-section oriented from southeast to northwest across the western Atlantic Ocean, New Jersey and eastern Pennsylvania (Seaman and Michelson, 2000). The figure is based on a 4-km MM5 simulation with multi-scale FDDA (analysis nudging and observation nudging) and reveals how the interactions of mesoscale vertical thermal and wind structures can influence pollution transport. In this case, 14 July 1995, the regional-scale low-level wind flow was from the southwest in a broad anticyclone, while winds aloft around 850-mb were weak and from the west (not shown). The mid-level westerly flow over the mountains induced an Appalachian Lee Trough (APLT) to the east, with its convergence zone over eastern PA (marked as WT in the cross-section). Since the low-level winds are southwesterly, they flow sequentially over Richmond, VA, Washington, DC, Baltimore, MD, and Philadelphia, PA before reaching the cross-section, and then continue toward northern NJ and New York City. A second convergence zone, marked by ET in the figure, lay directly over this urbanized corridor at the Delaware River. Upward vertical velocities are associated with the low-level convergence zones along the two troughs (not shown).

Notice that the mixed-layer depth in Fig. 4 is deepest at the inland troughs (about 1750 m AGL), but is more shallow in the modified marine air over NJ (about 500 m AGL) and is very shallow (about 60 m AGL) in the stable air east of the Atlantic Coast, marked AC in the figure. Meanwhile, a maximum in the upward vertical motion of 0.42 m s^{-1} occurs at the convergence zone of the eastern trough (ET) (not shown), with moderate

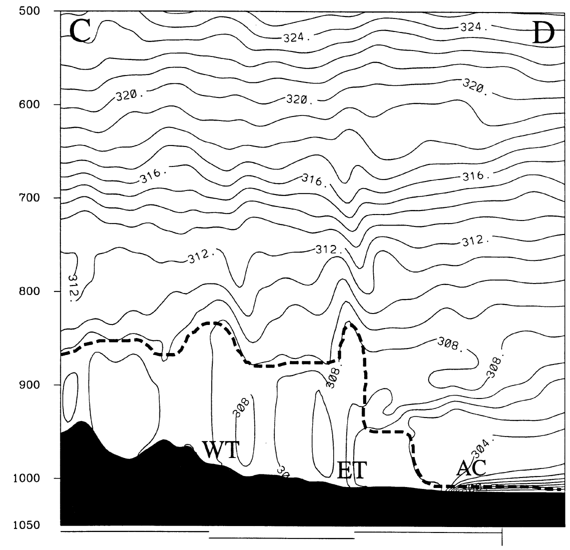


Fig. 4. MM5 cross-section showing potential temperature (K) and mixing depth (dashed line) at 1500 EST (2000 UTC), 14 July 1995. Isentrope interval is 1 K (thin solids). Point AC indicates Atlantic coast. Point ET (WT) marks the position of the eastern (western) branch of the Appalachian Lee Trough (from Seaman and Michelson, 1999, 2000).

westerly winds at 875 mb just east of the trough. Near the 875-mb level in the figure, the isentropes reveal an elevated mixed layer of warm air between 307–308°K that has been injected above the lower capping inversion over NJ (east of the eastern trough). Since the source of this elevated warm plume is the highly polluted air lofted in the deep mixed layer along the trough's convergence zone, the elevated warm plume is likely to have high concentrations of ozone and other contaminants. Once trapped well above the shallow coastal and ocean mixed layers, this elevated ozone plume was transported northeastward in a nocturnal low-level jet and reached New England the next day. Later that morning, it was mixed downward to the surface, causing ozone exceedances hundreds of kilometers from its source (Ray et al., 1998). This complex vertical structure, which is related to a combination of regional dynamics and boundary-layer turbulent processes, is an excellent example of the value of high-quality physical parameterizations in numerical meteorological models.

4. Requirements for reducing uncertainty in meteorological fields

Despite the evidence for comparatively lower errors in meteorological fields produced by DAMs, it is clear that remaining errors in their solutions can be significant. The principle sources of these errors can be divided into three categories: (1) data acquisition, analysis and assimilation, (2) model numerics, and (3) physical parameterizations. A fourth source of error affecting AQMs coupled with the meteorological models can occur depending on how those fields are used in the AQM. It is the objective of this section to review the current state of the science in each area, to identify the most important sources of errors and to suggest opportunities for reducing uncertainties in future applications. Unless stated otherwise, all subsequent discussions of modeling capabilities will refer to the dynamical models and DAMs.

4.1. Data acquisition, analysis and assimilation

The emergence of data-assimilating dynamical models as a viable tool for generating meteorological fields for air-quality applications in no way reduces the need for special observations. In fact, without a reasonably complete and accurate data base, DAM solutions converge to those of the non-assimilating dynamical models, with all their short-comings (Section 2.2). Since the end of World War II and the advent of practical weather radars, *remote sensing* of the atmosphere has become a critically important area of investigation impacting nearly every other area of meteorological research. While not replacing in situ measurements, remote sensing allows virtually continuous monitoring of atmospheric states aloft, either from the ground or from satellites. Some of the more important ground-based instruments which have become widely used are profilers, RASS, NEXRAD, SODAR, and LIDAR. Other sensing systems, such as the Automated Surface Observing System (ASOS) deployed by the US. Weather Service, allow measurement of a number of meteorological variables at the surface (wind, pressure, temperature, water vapor, precipitation, clouds cover, etc.). A full discussion of emerging atmospheric measurement technologies is given by Neff (2000).

The increasing availability of remotely sensed data and the computer technology to process it provide great opportunities for improving the quality of meteorological fields for air-quality applications. For some types of remotely sensed data, such as winds from radar profilers and SODARS, fairly reliable quality-control algorithms have been developed. Once they have been quality checked, these data can be assimilated in a straightforward manner. However, although the inflation-adjusted costs of deploying these instruments have declined, they remain substantial. Therefore, we must expect that these types of data will continue to be available only for limited areas during special field studies. On the other hand, environmental data from NEXRAD WSR-88D doppler radars and satellites are available almost continuously in many regions. Considering the huge volume of these data now available, one of the greatest problems to be faced in the years ahead will be to learn how to use these data more effectively, rather than considering ourselves as data-poor.

NEXRAD data are available from 132 sites in the US (roughly twice the number of twice-daily radiosonde sites). However, since NEXRAD reflectivity is not a meteorological state variable, it can be difficult to use in some DAMs. A great deal of research in the weather-forecasting community has been directed to better understand how to assimilate radar- and satellite-derived diabatic heating profiles (e.g., Tao et al., 1990; Manobianco et al., 1994; Chang and Holt, 1994; Lambert et al., 1994). These efforts, aimed at improving rainfall forecasts, have had limited success. Xu and Qiu (1994,1995) have developed an attractive application in which radar reflectivity and radial winds are used in a simplified form of the 4-D variational analysis (Section 2.3) to derive horizontal wind vector fields. This method appears to have considerable merit for recovering regional-scale winds (Porter et al., 2000) and will soon be tested in a DAM.

Another type of wind data routinely available from NEXRADs is the velocity azimuth display (VAD) wind profile derived from the doppler radar's radial velocity measurements. Since VAD winds represent an average over a radius of 50–60 km, they are very different from radiosonde or profiler soundings, which measure winds along a narrow path or cone. Michelson and Seaman (2000) have shown that these VAD wind data typically have low bias errors, but large standard deviation errors when compared to radiosonde winds. However, they developed a quality-control filter which allows the least-reliable data to be identified and discarded. When the filtered data were assimilated into a DAM, model-generated wind errors in the lowest 2.5 km were reduced significantly. Also, satellite-retrieved surface radiances can be used to estimate soil moisture and skin temperature, and McNider et al. (1994) have investigated assimilating these derived data in a DAM. Exploitation of these continuously available data types could prove extremely valuable for air-quality studies, especially those of long duration.

4.2. Model numerics

Another consequence of the rapid explosion in computational capacity is the widespread use of finer resolutions, larger domains and longer model integration periods in air-quality assessments. Through the 1980s, most meteorological modeling for multi-day applications were regional in scale, with grid resolutions of 20 km or more. Although a few 3-D real-case investigations were conducted at the mesobeta and mesogamma scales, most of them were run for shorter periods of about 6–24 h and used fairly small domains (e.g., Segal et al., 1988b; Seaman et al., 1989). On a limited basis, short-range studies in complex terrain were conducted with grid resolutions as fine as 0.5 km (Yamada and Bunker, 1988). It was recognized, however, that air pollution episodes lasting several days represented a multi-scale problem.

To better accommodate fine-mesh resolutions while simultaneously using long integration periods, nested-grid domains have become common in meteorological models. An advantage of nested-grid models is that they allow the lateral boundaries to be placed well away from the area of greatest importance. Failure to do so can lead to serious errors in the fine-mesh solutions, which may be dominated by the lateral boundary conditions (Yamada and Bunker, 1988; Warner et al., 1997).

As horizontal grid resolutions become finer, similar improvements for vertical resolution should be introduced. In general, the top of the meteorological model domain must be in the stable stratosphere (150 mb or higher) for correct representation of deep convection and jet-stream dynamics. In most air-quality applications, relatively high vertical resolution is essential in the PBL, while somewhat less resolution is acceptable in the upper atmosphere. As shown in Table 2, most recent modeling applications related to air-quality studies have used at least 30 layers and horizontal resolutions of 4-km or finer. In most cases, the lowest calculation level was placed no more than 50 m AGL. Based on the structure of observed and simulated capping inversions atop mixed layers over land (Shafran et al., 2000) and water (Leidner, 1995), it is likely that resolutions in mesoscale models on the order of 40–50 m throughout the boundary layer and capping inversion would be beneficial. This resolution may require a total of 50–60 layers in the vertical direction. To help offset the greater computational demands of finer-mesh models, older explicit time-differencing schemes, like the "leapfrog" scheme (Haltiner and Williams, 1980), generally have been replaced by more efficient implicit, split-implicit and split-explicit differencing methods (e.g., Pielke, 1984).

Further improvement of meteorological models also will require installation of more precise finite differencing methods. Most finite-difference schemes in use today already are either second-order or fourth-order accurate. Positive definite advection schemes or semi-Lagrangian differencing schemes have been tested in some models to reduce finite difference errors for the mass fields, especially in the vicinity of strong gradients (e.g., Smolarkiewicz, 1984; Smolarkiewicz and Clark, 1986). Bott (1989a,b,1993) refined the positive definite advection scheme, with nonlinear renormalization of the advective fluxes, to give excellent mass and gradient-conservation characteristics. Furthermore, it does not require extra horizontal diffusion to maintain numerical stability. Jakobs and Tilmes (1995) have tested the Bott advection scheme in a 3-D mesoscale meteorological model and found significant improvements in the gradients of water vapor along frontal bands. In their tests, Bott's scheme required only moderate additional computer time (about

15%), compared to a standard second-order advection scheme.

4.3. Physical parameterizations

Many physical processes critical to regional and larger-scale meteorology are fundamentally based on mechanisms rooted in the molecular scale and microscale. Since it is impractical to represent all of these processes explicitly in a mesoscale model, the sub-grid scale processes are represented in a simplified way. Written as sub-models within the parent dynamical modeling system, these *parameterizations* represent implicitly the bulk impact of the sub-grid scale physics on the resolved-scale environment.

Parameterizations generally contain assumptions and empirical relationships that allow complex processes to be calculated efficiently based on the resolved-scale variables and a set of equations describing the physical mechanism. Key *closure assumptions* are made to reduce the number of unknowns to equal the number of equations available to solve for them. The closure assumptions are critical to the accuracy of the sub-model and its range of applicability. For example, many parameterizations for representing deep convection (thunderstorms) assume that updrafts cover only a negligible fraction of any grid cell. This assumption becomes invalid for a mesh of 10 km or less, so the accuracy of these convective parameterizations is likely to deteriorate at finer scales.

4.3.1. Surface processes

Surface fluxes of heat, moisture, momentum and short/long wave radiation are crucial for air-quality applications because they are the primary mechanisms driving the development of the turbulent boundary layer. Land-surface parameterizations in nearly all advanced meteorological models are built around a prognostic energy budget equation for the earth's surface temperature, and they may include a prognostic equation for soil moisture, as well. Definitions of specific land-surface characteristics are needed for calculating the surface fluxes. These include, but may not limited to, albedo, roughness, thermal inertia, emissivity, vegetation height and type, leaf area index, plant water-reservoir capacity, wilting criteria, and the soil moisture and porosity in several sub-surface layers. In many regions, some of these data may be unavailable or only poorly known, and some are seasonally dependent. Therefore, it is common for surface sub-models to depend on assumed relationships between land use and the surface characteristics (e.g., Grell et al., 1994). The land-use types (anywhere from about 10 to perhaps 50 categories) usually are defined from satellite imagery or aircraft surveys of the land physiography (e.g., urban land, agricultural land, wetlands, etc.).

Many land-surface schemes of varying complexity are in use today. Among the more complete parameterizations are the Biosphere-Atmosphere Transfer Scheme (BATS, Dickinson et al., 1993) and the Parameterization for Land-Atmosphere-Cloud Exchange (PLACE, Wetzel and Boone, 1995). Not only do the more advanced sub-models predict the surface fluxes needed by the host atmospheric model, but they also predict the temperature, wind, humidity and water content within the plant canopy. These can be valuable for driving biogenic-emissions models. However, complex land-surface sub-models require definition of many parameters and variables for soils and vegetation. Although rapid progress is being made in defining certain of these values in the United States (e.g., Miller and White, 1998), many of the required inputs are not presently available at resolutions of 5 km or less for more remote regions. Most often, "typical" values are assigned to parameters associated with classes of vegetation (e.g., hardwood deciduous forests, grasslands) and soil types (e.g., sandy loams, clay), rather than relying on direct local measurements. This approach is an extension of the land-use paradigm and probably is quite reasonable for many cases. However, it can produce large uncertainties in the surface fluxes under some conditions (Wetzel and Boone, 1995). At the mesobeta scale, Wetzel et al. (1996) demonstrated that spatial variability of land-surface fluxes due to vegetation differences can affect the characteristics of cumulus cloud fields. Given the many degrees of freedom in these most complex land-surface schemes and the many parameters to be defined, their use to date has been limited mostly to basic research, rather than routine applications in regional models. A more detailed discussion of advanced land-surface schemes is provided by Dickinson (1995).

Less elaborate land-surface schemes normally are found in most mesoscale models used in air-quality studies. This class of simpler surface-flux sub-models often reduces the soil and vegetation parameters to a few that are found to be most important (e.g., Pleim and Xiu, 1995; Xue et al., 1991; Segal et al., 1988a; Avissar and Mahrer, 1988; Tremback and Kessler, 1985) or they may omit explicit plant canopies entirely (e.g., Zhang and Anthes, 1982; Noilhan and Planton, 1989). Although based on simplified methods, these schemes have been applied successfully to a wide range of applications meaningful for air-quality assessments. For example, Ulrickson (1992) has reported sensitivity of afternoon urban-scale circulations to surface characteristics and Copeland et al. (1996) have reported effects on regional climate due to vegetation changes, similar to the results of Seth and Giorgi (1996) using a more detailed surface scheme.

4.3.2. Turbulent processes

The study of turbulent processes is a very broad subject that cannot be treated in detail here. However, turbulent mixing and the so-called mixing depth are critical to the vertical transport of pollutants, horizontal plume dispersion and dry deposition in the PBL. This section briefly examines some of the principal approaches used to represent sub-grid scale turbulence in mesoscale meteorological models.

Boundary layer mixing is driven by thermal buoyancy and wind-shear. Diagnostic meteorology models often include a scheme to represent the mixing depth from the surface heat flux or temperature, a thermal sounding, and the wind shear (e.g., Berman et al., 1997). Most dynamical meteorology models used in turbulence sub-models have either a first-order K-theory closure (e.g., Noilhan and Planton, 1989), a non-local closure (Zhang and Anthes, 1982) or a simplified second-order closure (e.g., Mellor and Yamada, 1974,1982; Burk and Thompson, 1989; Ballard et al., 1991). The non-local closure usually is intended for convectively unstable conditions, while the first- and second-order closures can be applied to both stable and unstable conditions. The second-order schemes tend to be somewhat more computationally intensive, but generally have better performance characteristics than most non-local and first-order schemes (e.g., Shafran et al., 2000). While all three sub-model types are reasonably accurate in well-mixed conditions, they may have more trouble simulating the structure of stable nocturnal PBLs. Development of boundary layer schemes for stable conditions is an ongoing area of investigation (e.g., McNider et al., 1995).

In all of these parameterizations the scale of turbulent eddies is assumed to be well below the model's grid scale. This closure assumption is certainly true for models with meshes of 4 km or more. However, PBL depths can easily reach 2–3 km and if the grid resolution is 1 km or less, then the model will begin to resolve explicitly the largest, most energetic eddies (which scale to the PBL depth). Since the energy of the largest eddies can be represented twice in this case, serious errors can develop in the model solutions. Thus, turbulence parameterizations in very fine scale models must be modified to avoid this "double counting" of the larger eddies. Deardorff (1980) proposed a simplified second-order scheme with length scales that adjust to the grid size for unstable conditions, rather than being scaled only to the intrinsic turbulence scales. That is, the model grid is free to resolve whatever scales of motion it can (including the largest eddies), while only those eddy scales which are truly sub-grid are parameterized. This approach has been used with some success in cloud-scale models (e.g., Klemp and Wilhelmson, 1978) with grid-cell horizontal and vertical aspect ratios near 1 : 1. For large aspect ratios on order of 100 : 1, as found in regional and local models, a similar approach has been applied in the Advanced Regional Prediction System (Xue et al., 1996).

Despite the relative skill of second-order closure sub-models, compared to first-order and non-local schemes, they still may contain serious flaws. However, the kind of turbulence observations needed to improve these sub-models are difficult to obtain. An alternative to direct observations is to use turbulence fields generated in a high-resolution 3-D numerical model, called a large eddy simulation (LES). An LES typically has mesh sizes of less than 50 m and thus can resolve a range of eddies (e.g., Moeng, 1984). Using an LES data base, Moeng and Wyngaard (1989) found that second-order PBL schemes under-estimated the dissipation length scales, which led to underprediction of turbulent kinetic energy levels.

Otte and Wyngaard (1996) employed LES results to develop a PBL using Legendre polynomials as basis functions in a spectral framework. Their spectral PBL replicates LES results quite well and can be solved about as rapidly as first-order schemes. This sub-model has recently been installed in the 3-D MM5 mesoscale model and has performed well in early tests over the Great Plains and coastal regions (Stauffer et al., 1998). Such LES-calibrated spectral-framework turbulence schemes hold promise for reducing model errors, while limiting the need for extremely high vertical resolution in the PBL. Clearly, these advantages would be highly valuable for air-quality applications.

4.3.3. Soil hydrology

Soil moisture and evapotranspiration are closely linked (Wetzel and Chang, 1987) and together they have a large influence on PBL stability, mixing depth, cloud formation and rainfall. However, soil moisture is difficult to measure. Many schemes simply define the soil moisture as a climatological function of season and land-use type, or it may not change during the model integration, even if rainfall is predicted. Addition of even a simple time-dependent soil hydrology sub-model could significantly improve the accuracy of surface fluxes in multi-day, seasonal and annual model applications. For example, a land-surface sub-model, based on Pan and Mahrt (1987), has been adapted for use in NCEP's operational Meso-Eta model (Chen et al., 1997). This efficient and robust scheme includes canopy resistance and a soil-hydrology sub-model with surface runoff and ground-water recharge, allowing diurnal and seasonal evolution of soil moisture.

Both the scheme of Chen et al. (1997) and the more complex systems described in Section 4.3.1 require detailed initial moisture fields. Since 1997, NCEP has archived the Eta soil-moisture fields over the US. Otherwise, a soil-hydrology model can be run for a period of time (perhaps two or three months) to develop good quality spatially dependent soil-moisture fields (e.g., Capehart and Carlson, 1994; Smith et al., 1994). Regardless of the particular approach, soil-hydrology sub-models are likely to become an important element for reducing meteorological model uncertainties in future air-quality studies, particularly as seasonal and annual assessments become more common.

4.3.4. Radiation

The radiation flux divergence at the surface, or course, has a direct impact on the surface fluxes of heat and moisture. Radiation also plays an important role aloft, particularly when clouds are present. For short-period simulations, on order of a day or so, it may suffice to calculate the radiation budget only at the earth's surface, using a simple parameterization such as that of Zhang and Anthes (1982). However, in clear air, diurnally averaged radiation flux divergences lead to cooling rates on order of 1–2°C d^{-1}. Near cloud tops, radiative cooling easily can be larger by one to two orders of magnitude. Ignoring this cooling causes net warming of the column across the entire model domain through the other physical and dynamical processes (subsidence, diabatic heating and turbulent transport). In limited-area models, the net heating can lead to spurious domain-wide mass divergence and falling pressures. Therefore, for multi-day episodes a full-column radiation sub-model becomes fairly important.

Many column radiation parameterizations of different complexity appear in the literature and cannot be reviewed here. The more complex types represent the radiative spectrum as several different bands, but these schemes can be computationally expensive. Most global and regional-scale models use simpler, more efficient schemes (e.g., Slingo, 1980; Chen and Cotton, 1983,1987; Dudhia, 1989). In general, the minimum requirement acceptable for multi-day episodes is a full-column two-stream single-band radiation sub-model that includes the radiative effects of clouds.

4.3.5. Explicit moist physics and fog

Parameterizations also are required for the resolved-scale moist physics of clouds and precipitation (explicit moisture). When the grid-resolved water vapor reaches saturation, prognostic equations are activated for condensed water mass, with separate equations for various types of hydrometeors, depending on the complexity of the scheme (cloud water, pristine ice, rain, snow, graupel, hail). Advection of hydrometeors is treated explicitly, while other sub-grid terms represent conversion from one precipitation or cloud state to another (autoconversion, riming, sublimation). Thus, although referred to as "explicit" schemes, these resolved-scale cloud models are actually parameterizations themselves.

Explicit-moisture parameterizations range from fairly simple schemes that treat only warm clouds and precipitation (Hsie, 1984), to somewhat more complex systems that include simple ice-phase physics (Dudhia, 1989), and more elaborate systems with mixed-phase physics, additional ice-phase classes and interactions among the various hydrometeor types (Cotton et al., 1982,1986; Reisner et al., 1998). The more complete schemes are actually very similar to true cloud-scale models (e.g., Klemp and Wilhelmson, 1978). When applied at fine-grid resolutions (below 5 km), explicit moist-physics schemes also can simulate convection processes in mesoscale models.

Fog is another moist process acting within the boundary layer that can be important in cases with high-concentrations of particulate matter (PM) because of its influence on chemical reaction rates, secondary aerosol formation and deposition (Jacob et al., 1987,1989; Pandis and Seinfeld, 1989). In addition, fog affects the long- and short-wave radiation balance and may delay significantly the growth of the mixed-layer following sunrise. Delay of the mixed-layer growth can prolong high concentrations of emissions near the surface and change the evolution of the chemistry (e.g., Dye et al., 1999).

Fog is simply a stratus cloud at the ground, so most regional models currently represent it using the same explicit-moisture parameterization used for other resolved-scale cloud and precipitation processes. This approach, while straightforward, assumes that cloud or fog forms instantaneously and fills the grid volume as soon as saturation is attained. This can be appropriate for precipitation fogs and advection fogs. However, for radiation fogs associated with PM formation, patches and shallow layers often form in small valleys and depressions close to the ground (Holets and Swanson, 1981) and gradually grow upward to fill the grid volume. This process may not be resolved by the model's lowest layers and thus the simulated fog forms too late. Naturally, model skill for simulating fog formation also is strongly dependent on the other components of the water cycle, especially soil hydrology, evapotranspiration, vertical eddy transport in the boundary layer and rainfall. Some parameterizations based on the second-order PBL closure (Section 4.3.2) have been designed especially for representing fog development (e.g., Ballard et al., 1991; Musson-Genon, 1987; Gayno et al., 1994). Additional development and testing are necessary to take full advantage of the fog-simulation potential of regional models for air-quality applications.

4.3.6. Sub-grid shallow clouds

Shallow non-precipitating clouds (primarily cumulus and stratocumulus) do not produce significant precipitation and have little impact on the development of baroclinic storms. Consequently, they have been mostly ignored by the numerical forecasting community and generally are represented rather crudely in the models. For example, many models estimate only the area of sub-grid shallow cloud, based on the resolved-scale relative humidity (e.g., Benjamin, 1983; Slingo, 1987).

However, shallow-clouds can be important for understanding the vertical distribution of chemical species observed above cloud base (Ching and Alkezweeny, 1986). Processes important to the chemistry include the cloud-base vertical mass flux, cloud area, depth, radius, updraft velocity profiles, liquid-water distribution and entrainment-detrainment profiles. The updrafts can carry pollutants from the PBL into the free troposphere, where they are less subject to deposition and can be transported over greater distances. Wind speed and direction in the cloud layer can be significantly different than in the sub-cloud mixed layer, as well. Also, the mass flux in updrafts induces compensating subsidence in the environment, which affects the humidity and stability above the PBL. In addition these clouds can be thought of as moist reactor vessels affecting the chemistry. Because the simple relative-humidity cloud-diagnostic schemes cannot account for these processes, they are unsuitable for air-quality applications.

A number of schemes have been developed to represent the bulk effects of shallow clouds on their environment, primarily for use in global climate models. Some treat clouds as sites of enhanced mixing, so that the environmental thermodynamic and moisture profiles gradually adjust in the direction of the moist adiabatic lapse rate (Betts, 1986; Betts and Miller, 1986). More complete schemes calculate the vertical mass flux and use diffusion techniques to dissipate clouds gradually (Tiedtke, 1989,1993).

Meanwhile, observational research has led to development of process-based boundary-layer cloud models (e.g., Lilly, 1968; Wang, 1993). However, these mixed-layer and two-layer schemes are usually unsuitable for use in regional-scale meteorological models. Large eddy simulations (LES) have also been used in recent years to better understand the relationship between cloud properties, turbulence and the environment (e.g., Siebesma and Cuijpers, 1995; Bechtold and Cuijpers, 1995). An intercomparison of these different modeling approaches has been presented by Bechtold et al. (1996). Based on this work, Bechtold et al. (1995) have worked toward a unified approach for representing both shallow cumulus and stratocumulus in meteorological models.

Recently, there has been renewed interest in the air-quality community in the importance of shallow clouds to atmospheric chemistry, transport and mixing. Sub-models suitable for use in 3-D regional models and designed to represent the range of cloud processes needed for chemical applications include those of Walcek (1993), Seaman et al. (1996) and Kain et al. (1996). While this work represents a beginning, considerable model development and evaluation are still required.

4.3.7. Deep convection

Since ozone production in North America is dominated by photochemistry, which is favored by hot temperatures and clear skies, it sometimes has been assumed that rainfall can be ignored. While this assumption may be justified in especially dry climates (e.g., Los, Angeles), it is unreasonable for more humid environments. For example, high-ozone episodes in the Eastern US often have deep, moist unstable boundary layers, considerable convective available potential energy (CAPE) and only modest stable layers to inhibit deep convection. The high boundary-layer humidities not only favor deep convection, but actively contribute to secondary aerosol and haze formation. These high-ozone cases tend to be regional in scale and multi-day in length, so it is virtually certain that they will be accompanied by embedded clusters of thunderstorms.

Deep convection also can be a major factor toward improving air quality at the surface because much of the unstable polluted boundary-layer air can be drawn into rapidly growing convective updrafts, which carry it into the upper troposphere. This boundary-layer air is replaced by relatively clean moist downdrafts originating from the mid-troposphere. In addition to this vertical redistribution, wet deposition to the surface can be a significant removal mechanism, particularly for the more soluble species.

Most convective parameterizations assume that the area of an updraft is small compared to the area of a grid cell. The scale of typical thunderstorm updrafts varies from about 2–5 km (larger for supercells). Weisman et al. (1997) have shown that for a grid mesh finer than about 5 km, it is arguable that a model's explicit moist-physics can simulate the convection adequately. However, they also found that when coarser grids were used, convection developed and moved too slowly. Thus, for models with larger grids a convective parameterization is required. Wang and Seaman (1997) showed that several parameterizations can be effective in most situations for a mesh as fine as 12 km. For the range of about 6–10 km, however, the crucial scaling assumptions are violated for most of the parameterizations (Molinari and Dudek, 1992).

Many convective schemes have been developed for regional-scale and large-scale meteorological models and a review is given by Molinari and Dudek (1992). Among the most widely used are the Anthes-Kuo (Anthes, 1977); Betts-Miller (1986), Grell (1993) and Kain-Fritsch (1990); Kain-Fritsch (1993) parameterizations. The Anthes-Kuo and Betts-Miller schemes were designed primarily for coarse grids and in their original form do not include moist downdrafts. The Grell and Kain-Fritsch schemes directly include moist downdrafts, and so are better suited for simulating the development of cold pools, outflow boundaries, mesohighs and mesolows which accompany thunderstorms and MCSs. None of these schemes is recommended for use below 10 km.

4.4. Compatibility with air-quality models

4.4.1. Interfaces between models

In recent years it has been found that the sources of serious meteorological error in air-quality models are not limited to the meteorology-generating models alone (e.g., Hariharan and Venkatram, 1996; Pai et al., 1998). Byun (1999a,b) has provided an excellent discussion of the causes of errors due to inconsistencies between the two types of models. A number of problems can arise because AQMs and meteorology models have developed independently, for the most part, and therefore they generally require an interface or "connector" program. Key factors which may need to be reconciled by the interface program are the horizontal mesh sizes, vertical coordinate systems, the number and spacing of the vertical layers, and the map projections. If the grids of the two models are incompatible, an interpolation step is necessary to project the meteorology onto the AQM's grid system. Interpolation from one grid to another can lead to a change of mass and loss of detail, especially if the original grid has greater resolution than the target grid. These effects can be particularly important where strong vertical gradients exist, as in temperature and moisture at inversions, or in low-level jets.

Another problem involves divergences in the horizontal wind field. Since it is about five orders of magnitude smaller than the wind, small interpolation errors can cause much larger errors in the divergence. Thus, if an AQM diagnoses vertical velocity from the divergence, instead of using the meteorological model's vertical velocities directly, the inconsistency can produce large errors in those diagnosed velocities. Even if the meteorological model's vertical winds are used, faulty divergences caused by interpolations can have disastrous effects on chemical concentrations. As with diagnostic models (Section 2.1), the response often has been to apply a mass-consistency scheme following the interpolation. However, this step can lead to inappropriate changes to the vertical velocities. Thus, interpolations should be avoided whenever possible. Finally, compatible map scales are preferable and a number of models have multiple options. The most common are Lambert conformal, polar stereographic and Mercator.

4.4.2. *Use of meteorology within air-quality models*

Additional issues arise due to the application of meteorological fields within the AQMs. These issues may vary from model to model, but several types are common.

First, there is the issue of temporal consistency. In virtually all applications to date, the AQM is run subsequent to completion of a meteorological simulation. In an episode lasting several days, the AQM requires many hundreds of time steps, usually at intervals of several minutes. Normally, it has been infeasible to store the meteorological fields that often, so the AQM must perform a temporal interpolation to estimate the meteorology at the intermediate time steps. Some AQMs even hold the meteorology constant for an hour (or for whatever period is dictated by the input) and then update it instantaneously at the end of that period. Clearly, these temporal interpolations can lead to sizable mass errors. Byun (1999b) has suggested a mass correction scheme inside the AQM to reduce errors due to temporal interpolations. Vogel et al. (1995) eliminated such interpolations by coupling the meteorology and chemistry models at every time step.

Second, there is concern that non-hydrostatic pressure perturbations in the meteorological model can be incompatible with a hydrostatic assumption made in the AQM. That is, an AQM may assume that the air density at any level is a function of the vertical gradient of pressure through the hydrostatic equation. If the pressures from the non-hydrostatic model are integrated to obtain density (or some similar relationship based on hydrostatic balance), then errors can be introduced. This difficulty arises at the basic level of the dynamic meteorological equations, as discussed by Byun (1999a,b). It was suggested that the primitive meteorological equations be written in a mass-conservative form to ensure greater compatibility with the AQMs.

Third, there is often incomplete or inconsistent meteorological coupling between the two models at the parameterization level. That is, the physical assumptions in the meterological model may be different than those used in the AQM. Many examples could be cited. However, they generally occur because either some information needed by the AQM was not calculated by the meteorological model, or else it was calculated and not saved. For example, the actinic flux (solar energy at wavelengths active in photochemistry) is needed by many AQMs and is highly dependent on the distribution of clouds. Yet many mesoscale meteorological models assume that clouds exist only when a grid cell becomes saturated (i.e., an explicit cloud). Few meteorological models currently calculate actinic flux, since it is not used for the model's own integration. Thus, the actinic flux has to be estimated in the AQM from whatever meteorological variables are available. However, the cloud diagnosed in the AQM may be quite different from the explicit cloud predicted in the meteorological model unless the full moist physics is carried into the AQM. A meaningful study would be to calculate and verify the actinic flux within the meteorological model, rather than diagnosing this variable in the AQM.

Another area of potential inconsistency is in the treatment of convective precipitation. Many meteorological models have sophisticated deep-convection physics that interact with the resolved scales in many ways (e.g., Kain and Fritsch, 1993). In most AQMs, however, important sub-grid scale physical processes associated with deep convection, such as rapid vertical transport, may be neglected or represented poorly. McHenry et al. (1996) have shown that ensuring better coupling of the physics by introducing convective-parameterization modules from the meteorological model into an AQM can result in improved accuracy for the AQM chemistry.

A partial answer to these problems is to give more attention to which meteorological variables are saved and how those variables are used later in the AQM. However, it is important to work toward compatibility in the basic numerics of the two models, as well. Model compatibility must be an ongoing process as the models evolve, so that improvements in one model do not create new incompatibilities in the other. Compatibility issues can become quite complex in model-intercomparison projects, such as the Cooperative Regional Model Evaluation (CReME) study, where two or more meteorological models were used to provide inputs for different AQMs (Hanna et al., 1996; Fernau and Pai, 1998). Better documentation of significant changes to the codes is often helpful to prevent the development of new incompatibility errors.

In the final analysis, it may be best to develop fully coupled meteorological-chemical modeling systems, as done by Vogel et al. (1995). In this way fresh meteorological data are available to the chemistry modules at every time step, consistency can be attained at the internal level and no quantities have to be re-diagnosed. It should be mentioned that simultaneous integration of the two models does not require that the chemistry feed back to the meteorology (two-way interactive models), since the chemistry has only very minor impact on meteorological variables in most cases (Russell and Dennis, 2000). Rapid reductions in the cost of computer memory and high-performance processor chips are making this type of integrated modeling system more feasible.

5. Future investigations and developments

5.1. *Model evaluation and uncertainty assessment*

Clearly, the socio-economic implications of imposing air-quality standards and emissions limits are enormous for both public and private stakeholders. Given these circumstances it is important that the scientific guidance provided to policy makers undergo thorough scrutiny. Reliance solely on expert opinion, however valuable, is likely to be unsatisfactory.

Consequently, all types of modeling systems used for air-quality assessments, including meteorological processors, undergo repeated evaluations and uncertainty assessments. That effort is probably more advanced in the air-chemistry and emissions communities than it is in the meteorological community in general, because such a large part of meteorological research over the past 50 yr has been devoted to improving forecasts of variables that are poorly related to the needs of air-quality studies. As mentioned earlier, the weak-dynamics cases of greatest interest for air-quality studies (stagnant, hot anticyclones with little rain) tend to be the very cases of least interest to weather forecasters. Thus, meteorological model evaluation has tended to focus on techniques more appropriate for cases with strong dynamic forcing.

Evaluation of meteorological processors for air-quality assessments has been a subject addressed at a number of workshops and meetings. However, no standard protocol for evaluating model skill has emerged that is widely recognized in the air-quality field. This is unfortunate, because it is often difficult to compare performance among various models and in different environments. Consequently, it can be difficult to measure long-term progress as improvements gradually are introduced. Moreover, the absence of a standard evaluation protocol complicates the task of policy makers who need to assess the credibility of model applications. Therefore, it is recommended that interested stakeholders and air-quality scientists establish a set of minimum model-evaluation standards and protocols for meteorological processors. Establishment of such a minimum standard and enforcement by the agencies sponsoring air-quality research would be a significant step toward improving our understanding of meteorological model uncertainty, which would benefit both public policy and scientific inquiry.

A wide variety of statistics have been found to be useful (e.g., Anthes, 1983; Willmott, 1982; Willmott et al., 1985). However, a core set of statistics is suggested for routine use in air-quality applications:

(1) vector mean wind speed error for surface winds (hourly),
(2) mean speed error for surface winds (hourly),
(3) mean direction error for surface winds (hourly),
(4) mean error for surface temperature (hourly),
(5) mean error for surface mixing ratio (hourly),
(6) root mean square error for surface wind speed (daily and at selected times),
(7) root mean square error for surface temperatures (daily and at selected times),
(8) root mean square error for surface mixing ratio (daily and at selected times),
(9) mean error of mixing depth (selected times),
(10) root mean square error for mixing depth (selected times),
(11) index of agreement for surface wind (daily) (Willmott, 1982),
(12) scatterplots of observed versus model-generated surface temperatures (maxima, minima, daily),
(13) scatterplots of observed versus model-generated surface wind speeds (daily),
(14) scatterplots of observed versus model-generated surface mixing ratios (daily),
(15) vertical profiles of mean wind speed error, level by level (hourly or daily),
(16) vertical profiles of mean wind direction error, level by level (hourly or daily),

(17) vertical profiles of mean temperature error, level by level (hourly or daily),
(18) vertical profiles of root mean square wind speed error, level by level (hourly or daily),
(19) vertical profiles of root mean square wind direction error, level by level (hourly or daily),
(20) vertical profiles of root mean square temperature error, level by level (hourly or daily).

Additional statistical quantities can be helpful in certain cases. For example, if tracer measurements are available, mean plume dispersion can be evaluated, as well (Fast, 1995; Mueller et al., 1996). It is helpful if these quantities can be calculated at intervals following the tracer release, depending on data availability. Tracer studies can have a number of significant problems, however, (e.g., it is often difficult to account for all of the tracer material using the measurements), so great care must be taken in their design. Finally, it should be noted that important mesoscale meteorological features may exist on only parts of a domain (e.g., a sea breeze), so it may be necessary to obtain statistics on one or more sub-regions, in addition to generating domain-wide statistics.

5.2. New applications

Certainly, the future direction of air-quality management strategies will continue to evolve along with our understanding of the health and environmental impacts of air pollution. In addition to the suggested model improvements discussed in Section 4, there are a number of new issues raised by changing policy and the limited availability of research funds. While all the implications of what is being learned cannot be fully anticipated, of course, several concepts have emerged to help guide the development of meteorological processors in the years ahead.

For example, with better understanding of the adverse health effects due to long-term exposure to ozone and particulates, there has been a noticeable shift of interest toward the study of seasonal and annual air-quality characteristics and inter-regional transport. At present, it is not clear that existing regional-scale meteorological models, which have been carefully evaluated for shorter episodic studies, also are optimal for these longer periods. When model products are needed for such lengthy intervals, they often are generated in segments of 5–10 d. However, FDDA techniques make it feasible, in principle, to run a DAM continuously for a year or more. It is unknown which is the best way to perform such long numerical integrations. Direct comparison of these alternative approaches has not appeared in the literature, so far as is known.

For local air-quality considerations, it is often assumed that turbulent mixing is the dominant meteorological process because of its short time scale. Over longer periods and inter-regional scales, however, the cumulative effects of more slowly-acting processes, such as clouds, radiation and precipitation, have time to exert important influences. Most North American inter-regional transport studies were performed at least a decade ago, before FDDA systems were widely available, and with comparatively coarse grid meshes (e.g., Haagenson et al., 1987,1990; Chock and Kuo, 1990). Thus, the effectiveness of current-generation models for long-range (inter-regional) transport needs to be re-evaluated, perhaps using tracer-experiment data, and the sources of long-term biases in the regional model solutions need to be identified and corrected.

Moreover, better coupling of meteorological and air-chemistry models is certain to take place. To date, most applications have focused on creating a single “best” set of meteorology, followed by numerous chemistry simulations in which emissions or internal reaction mechanisms can be altered. The limitation of computer resources has been the primary reason for this approach, but it has had a number of negative consequences (Section 4.4). Fully integrated meteorological and air-chemistry models can better address these issues and should have significant impacts on model skill in the future.

Another area certain to develop in the next few years is related to real-time forecasting of air quality using deterministic numerical models. A number of groups in the US and Europe already have begun experimentation on a limited basis. Given the known health impacts of airborne pollutants, it is remarkable that routine numerical air-quality forecasts were not initiated at least a decade ago. By contrast, numerical weather prediction research was begun soon after the birth of the computer age (Charney et al., 1950), so that by the late 1950s, routine numerical weather predictions were being run operationally by the US. Weather Service. Although model skill initially was below that of experienced humans, operational use greatly accelerated model development. Today, even the most skilled weather forecasters feel at a tremendous disadvantage if a numerical prediction is unavailable.

Similar moves to exploit Eulerian air-chemistry modeling systems for predictive purposes have been slow to develop. Only recently has the US. National Research Council recommended that the National Weather Service consider providing more of the specific meteorological variables necessary for real-time air quality forecasting. No doubt, uncertainties about atmospheric chemistry and the absence of detailed 3-D measurements for model initialization have played an important role in causing the long delay. However, improved monitoring of emissions and airborne pollutants, plus better understanding of atmospheric reaction mechanisms, has eased this situation considerably. The emergence of regular real-time air-chemistry forecasts, although initially used primarily for research, can be expected to stimulate significant improvements in model skill and to bring real benefits for public health and welfare.

6. Conclusions

Meteorological fields are required as inputs for air-quality models, but they often contain significant errors which, in turn, contribute to errors in simulations of airborne chemical species, aerosols and particulate matter. Atmospheric states can be diagnosed from observations or simulated by dynamical models (with or without FDDA). In general, diagnostic models based on observations have an advantage in that they represent actual atmospheric states and the data reflect the influence of all scales of motion and physical processes affecting the measurement sites. However, direct measurements are costly to obtain, may omit key variables, and often lack sufficient spatial or temporal density to describe the fields adequately. Also, diagnostic models lack dynamic consistency among the variable fields because they are not based on the complete primitive equations. Nevertheless, they remain important for some types of studies for which adequate data are available and where mesoscale assessments are needed either very rapidly or for lengthy periods of several years.

Dynamical models, although far from perfect, have improved dramatically in recent years. With the introduction of FDDA techniques (DAMs), non-hydrostatic equation frameworks and improved physical parameterizations, they have become widely accepted for air-quality model applications in cases where observations alone are inadequate to define characteristics of the key meteorological fields (e.g., winds, temperatures, mixing depths, water vapor). Introduction of faster computers already allows simulations lasting up to 5–10 d on regional-scale domains and with resolutions of 1 km or less. At the same time, improvement of remote sensing technology is making new data sources available, such as NEXRAD WSR-88D winds and reflectivities. Development of new assimilation techniques now allows these data to be used in DAMs.

Dynamical meteorological models and DAMs are very complex systems which must continue to develop in the future. Publication of new technical approaches and application results are very important parts of model evaluation, but alone they are insufficient to fully understand performance. Therefore, it is strongly recommended that all models used in air-quality assessments be available in the public domain so they can undergo thorough independent scrutiny. This is an important policy decision necessary to stimulate scientific development and ensure availability of the best guidance based on tools that are clearly understood and recognized by the scientific peer community.

Based on this review of current state-of-the-science models and projected demands for meteorological information for future air-quality assessments, the following major conclusions are made:

(1) For most purposes, data-assimilating dynamical models are currently the best method available for generating reasonably accurate and dynamically consistent meteorological fields required for air-quality applications. Furthermore, they have the greatest potential for improvements in skill over the next several years.
(2) The rapid expansion of remote sensing technology provides an outstanding opportunity to reduce errors in meteorological fields, but research is needed to learn how to best use these data in analysis and assimilation techniques.
(3) As better land-surface schemes are introduced into meteorological models, considerable benefit may be gained by better coupling with emissions models, perhaps leading to fully integrated meteorological-emissions modeling systems.
(4) Rapid growth in computer technology has made it feasible to run multi-day regional meteorological-model simulations having very fine grid resolutions. To take full advantage of this new technology, however, more research is needed to develop scale-appropriate physical parameterizations (land/surface fluxes, soil hydrology, boundary layer fluxes, deep convection, shallow clouds).
(5) Mandated limits on long-term exposure to pollutants will require greater emphasis on long-range transport. This necessitates re-evaluation of mesoscale models for inter-regional transport problems, particularly in episodes of poor air quality associated with weak dynamical forcing and convection.
(6) Stakeholders should press for the adoption of a minimum set of standard model-evaluation procedures, based on available statistical tools, to allow more objective comparisons among various modeling systems and their applications.
(7) Better coupling and interfacing of meterological models and AQMs is a significant need, and preliminary efforts are being made to reduce errors due to model incompatibilities. Eventually, this should lead to fully integrated models that perform both meteorology and chemistry processing.

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