

Numerical Air Quality Modelling: Past, Present and Future

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Introduction

In this presentation, an overview will be given of the recent history, current status and future direction of three-dimensional numerical air quality models. Particular emphasis will be given to photochemical air quality simulation systems (PAQMS), because these are now widely used both in Australia and Internationally. In considering the development of such modelling systems, it is noted that their evolution has seen three major stages of development. A brief description of each development stage will be provided.

Definitions

Photochemical air quality simulation systems are based on the semi-empirical transport-diffusion equation, which, in a Cartesian framework, has the following form.

$$\frac{\partial C_i}{\partial t} + \tilde{\nabla} \cdot (\tilde{u} C_i) = \nabla \cdot \tilde{K} \cdot \tilde{\nabla} C_i + R_i + Q_i \quad , i = 1, \dots, n \quad (1)$$

where C_i is the volume averaged, ensemble mean concentration of species i , \tilde{u} is the time and volume averaged 3-dimensional wind vector, \tilde{K} is the volume averaged eddy diffusivity tensor, R_i is the ensemble and volume averaged rate of chemical transformation of species i , and Q_i is a source term. The solution of (1) is achieved through defining a computational mesh covering the region of interest and integrating the governing equations over the period of interest (generally for 2–7 days). Typically (1) is split into a number of 1-dimensional analogues which are solved by numerical algorithms which are specifically tailored for the process being modelled (i.e. advection/diffusion/chemical transformation). In order to fully define (1), it is also necessary to prescribe time-varying 3-dimensional meteorological fields, an inventory of emissions for the relevant species (in the case of photochemical smog, emissions of oxides of nitrogen, carbon monoxide and volatile organic compounds).

A schematic of a photochemical air quality modelling system (PAQMS) is given in Fig. 1. It can be seen that the system includes a meteorological transport model, an emissions inventory module and a chemical transport model. If the meteorological model is integrated prior to the chemical transport model integration then the chemical transport modelling is said to be undertaken in an offline mode (see Stage-I and Stage-II model development below). If the chemical transport and meteorological process are integrated within the same time loop then the chemical transport modelling is said to be undertaken in an online mode (see Stage-III) modelling.

We now go on to describe the three stages of evolution of photochemical air quality modelling systems.

First generation systems

First-generation PAQMS were conceived in the U.S. in order to provide a tool which could be used to investigate control options as required by the 1970 Clear Air Act (Scheffe and Morris, 1993). First-generation PAQMS were limited by small computer capacity, by a limited experimental data base, by a limited mechanistic understanding of the photochemical transformation process, and by little, or poor quality emission data bases.

High quality three-dimensional meteorological data sets were also lacking at this time. Because of these constraints, the systems were applied to relatively small domains, and the photochemical transformation mechanisms were highly condensed. For example, a first-generation PAQMS was developed by Reynolds et al. (1973, 1974) in order to investigate the occurrence of ozone in the Los Angeles region. The model considered the chemical transformation of 11 species via 15 reaction pathways. Six of the 11 species were transported and the remainder treated as steady state. The model was applied over a 25 x 25 x 5 grid with a horizontal resolution of 3.2 km, and a vertical resolution that varied with the mixed-layer height. Simulations were conducted for six Los Angeles days (selected from the autumn of 1969). The model was evaluated against ground-level observations of carbon monoxide (CO), nitric oxide (NO), nitrogen dioxide (NO₂), total hydrocarbon, and ozone.

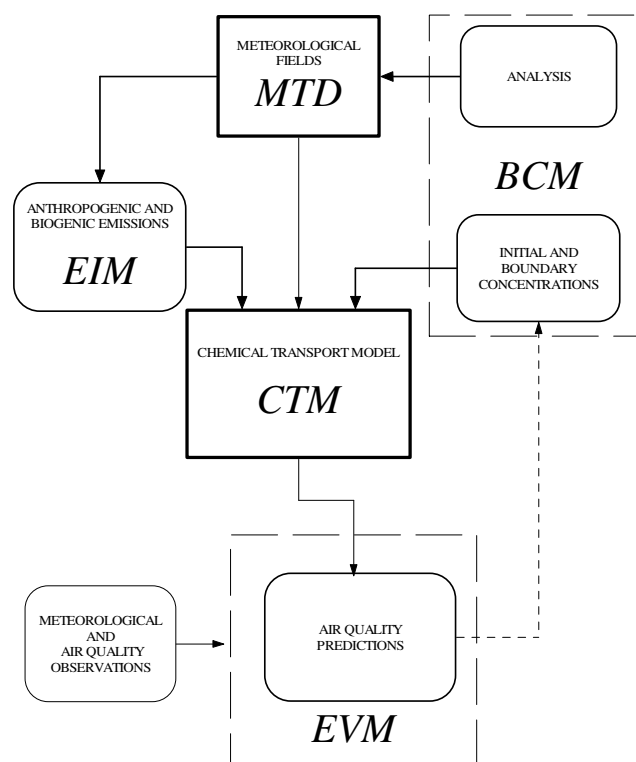


Fig. 1. Schematic diagram showing the components of a Photochemical Air Quality Modelling System (PAQMS). CTM- chemical transport model; MTD- meteorological transport driver; EIM- emissions inventory module; BCM- boundary condition module; EVM- evaluation module.

Second generation systems

The second generation of PAQMS featured multi-scale models. In the U.S. and Europe, it was recognised that inter-regional transport of pollutants played an important role in defining the spatial and temporal distributions of photochemical smog over urban areas. To this end, models were developed which incorporated multiple one-way nesting in the horizontal, or variable resolution grids. An example of such a model is the Urban and Regional Multiscale (URM) model (Kumar and Russell 1996) which uses a two-dimensional finite element mesh with variable size 'bricks'. When modelling ozone production over the eastern U.S. high resolution is prescribed over the urban areas (4.626 km), and low resolution is prescribed over the rural areas (18.5–74 km).

The second generation modelling systems were also notable in that they incorporated the effects of volatile organic carbon emissions from biogenic sources- via meteorologically dependent emission models (Guenther et al., 1991), and via extensions to chemical transformation mechanisms to deal with the oxidation of

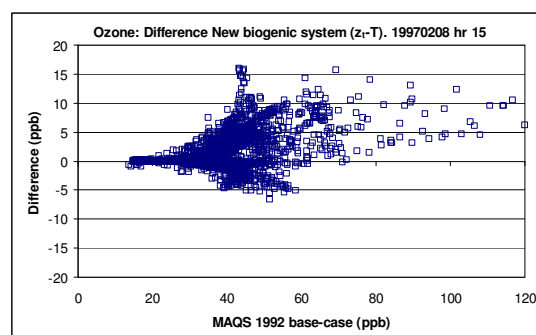


Fig. 2. Concentration difference scatter plot for a modelled Sydney ozone day (hour 16 on 8 February 1997), showing the sensitivity of 1-hour ground-level ozone concentrations to biogenic VOC fluxes. Each point of the scatter plot corresponds to a ground-level cell in the model and shows the difference in the predicted 1 h ozone concentration resulting from including biogenic emissions in the emissions inventory.

species such as isoprene and mono-terpenes (Carter 1996). The effect of including biogenic emissions is illustrated in Fig. 2 for a modelled Sydney photochemical smog event (CSIRO, 2002). It can be seen that biogenic emissions are predicted to increase ozone concentrations by up to 15 ppb.

Although diagnostic meteorological models were still used to drive second generation models, prognostic meteorological models utilising four-dimensional data assimilation became the primary meteorological drivers of PAQMS. Note however, that the systems were run in an offline mode. Second generation PAQMS have also included plume-in-grid methods for explicitly tracking point source emissions until plume dimensions became comparable to the grid size of the three-dimensional model. The plume mass was then added into the host model. Advanced numerical solution procedures were also developed specifically for PAQMS. This included highly accurate one dimensional and two dimensional advection schemes (i.e. Yamartino 1993) and pragmatic single step chemical integration methodologies.

Supporting observation data sets for verifying second generation models also increased in complexity. It was recognised that model solutions were non-unique, and thus could be generated through processes in which compensating errors may lead to seemingly correct solutions. As a consequence, considerable resources have been put into ensuring that individual processes in modelling systems are adequately verified, including the meteorological fields, the emissions inventories and the chemical transformation mechanisms. Verification of chemical transformation required the measurement of speciated volatile organic compounds (VOC) and oxides of nitrogen concentrations within urban areas, the measurement of biogenic species and oxidation products downwind, and the measurement of a suite of photochemical smog products (including hydrogen peroxide, nitric acid, organic nitrates, in addition to ozone). In the U.S., specific sites (PAMS-<http://www.epa.gov/air/oaqps/pams/>) have been set up to measure photochemical precursor and product species on a routine basis. Diagnostic models have been built which relate the ratios of key photochemical species to the regime of photochemical development (Sillman 2000). Second generation photochemical modelling systems have been used extensively both in Australia and internationally, principally addressing issues relating to the development of long-term control strategies for the mitigation of photochemical smog.

Third generation systems

Third generation systems are now becoming routinely available (Byun and Ching 1999; Hurley 2003). Such systems may include comprehensive sub-models of aerosol processes, catering for both physical (condensation, nucleation, and coagulation) and chemical aerosol processes. Moreover, although some third generation systems are run in an offline mode, increasingly, chemical/aerosol transport/transformation is simulated in an online mode, in which the chemical and aerosol processes are integrated within the main time marching loop of the meteorological model. Such an approach is desirable because it enables more frequent updates of the meteorological fields (in principle providing more accuracy), and because it provides the potential for feedback between the trace gas and aerosol concentration fields and the radiative forcing and cloud dynamics in the meteorological model.

It is notable that the significant mass conservation constraints imposed by the use of online chemical/aerosol schemes within numerical weather prediction systems, has promoted the development of numerical weather prediction systems which use mass conserving forms of the governing differential equations (i.e. <http://www.mmm.ucar.edu/wrf/users/document.html>).

Third generation models are also multi-scale. However while this is generally achieved through fixed one- or two-way grid nesting techniques, researchers are also considering adaptive grids, which concentrate node density in areas of strong spatial gradients of the prognostic meteorological or pollutant fields (Srivastava et al., 2000). The numerical solution of chemical and aerosol mechanisms is also an area of on-going research, in which Rosenbrock techniques (Verwer et al., 1999) show considerable promise.

Third generation three-dimensional numerical grid models are being used to look at photochemical and coupled photochemical-aerosol problems, over scales which range from sub-urban (i.e. 1 km grid size) to continental. On the urban scale, boundary-layer modelling is the subject of increasing attention, with consideration given to correctly describing urban energy, moisture and momentum budgets. Third generation numerical modelling systems are also increasingly being used for air quality forecasting (Cope et al., 2004). For airsheds which exhibit high levels of persistence, the accurate prescription, and assimilation of initial concentration fields is critical to good forecast performance. To this end, work is underway on methods for assimilating observed pollutant concentration fields into the models. Methods under consideration include the use of adjoint solutions, ensemble approaches and Kalman filtering (Daescu and Carmichael 2003).

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