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Evaluation of the Equilibrium, Dynamic, and Hybrid Aerosol Modeling Approaches

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The continued development of the dynamic and hybrid approaches (Pilinis et al. 2000; Capaldo et al. 2000) for the simulation of atmospheric aerosol dynamics is discussed in this paper. A linear interpolation method is proposed for the mapping of the moving aerosol size/composition distribution onto a fixed size grid. The 3 aerosol modules are incorporated into a trajectory model that includes descriptions of gas-phase chemistry, secondary organic aerosol formation, vertical dispersion, dry deposition, and emissions. The 3 approaches are evaluated against measurements from the Southern California Air Quality Study (SCAQS). All 3 models predict the 4–6 h averaged $PM_{2.5}$ (particulate matter with diameter $\leq$2.5 microns) and PM_{10} (particulate matter with diameter $\leq$10 microns) mass concentrations of the major aerosol species with errors $<$30%. For the aerosol size/composition distribution, however, the dynamic and hybrid models show better agreement with measurements than the equilibrium model. The hybrid model aerosol size distribution predictions are similar to the dynamic model results. The hybrid approach in this case combines accuracy with computational efficiency. The dynamic approach is the most accurate, but at a higher computational cost. Daily average $PM_{2.5}$ aerosol species predicted by the aerosol models with 16, 8, and 4 size sections all give reasonable agreement with the measurements. All 3 aerosol models show consistent sensitivities of nitrate, sulfate, and total $PM_{2.5}$ to the changes of NO_x, VOCs, NH_3, and primary sulfate emissions.

Keywords air quality model, aerosol dynamics, linear interpolation, $PM_{2.5}$, PM_{10}

INTRODUCTION

Atmospheric aerosols are multicomponent mixtures of sulfate, nitrate, chloride, ammonium, sodium, trace metals, carbonaceous material, crustal elements, and water. It is now recognized that these particles have adverse effects on human health and visibility and play an important role in climate changes. Understanding the relationship between emission changes and ambient concentrations of atmospheric aerosols is necessary for the establishment of effective control strategies to reduce their adverse impacts.

Mathematical models are often used to develop such relationships. However, detailed modeling of the aerosol dynamics is demanding because of the wide aerosol size range spanning diameters from a few nanometers to tens of microns. Because the mass transfer rate between gas and aerosol phases is strongly dependent on aerosol size, the mass transfer rates for the smallest and the largest particles can be different by several orders of magnitude. The mathematical description of such systems results in a set of extremely stiff differential equations.

The first generation of aerosol models employed equilibrium approaches (Pilinis et al. 1987; Russell et al. 1988; Binkowski and Shankar 1995; Lurmann et al. 1997). These studies assumed instantaneous mass transfer between the gas and aerosol phases with the composition of the latter determined by thermodynamic equilibrium. The major advantages of the equilibrium methods are their speed, simplicity, and stability. It has been shown, however, that under certain conditions, equilibrium cannot be established over the atmospheric relevant timescales of minutes to a few hours (Wexler and Seinfeld 1990; Meng and Seinfeld 1996). To address this problem, dynamic methods have been developed (Meng and Seinfeld 1996; Meng et al. 1998; Jacobson et al. 1996; Jacobson 1997a, b; Sun and Wexler 1998a, b) to better simulate mass transfer between the gas and aerosol phases. However, the excessive computational cost required by these dynamic models has led these investigators to employ certain restrictive simplifications, such as electroneutrality constraints on the species flux (Sun and Wexler 1998a, b).

Recently, Pilinis et al. (2000) proposed a more general approach that allows changes in aerosol acidity while maintaining the stability of the system. In spite of these efforts, dynamic models are often limited in their applicability to large-scale

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air-quality models by their computational cost. To combine the accuracy of dynamic methods with the efficiency of equilibrium methods, a hybrid approach was proposed (Capaldo et al. 2000). In the hybrid method, the condensation/evaporation of aerosol particles with diameters less than a threshold diameter (around 1 μm) are simulated using the equilibrium method, and the dynamic approach is used for the larger particles. This hybrid method has been used only in a box model, however, and has not been evaluated against ambient data. These 2 models did not include descriptions of coagulation and nucleation.

The moving sectional method (Gelbard 1990; Kim and Seinfeld 1990), where the boundaries of each aerosol size section move with time in accordance with the growth or shrinkage of aerosol particles at those boundaries, has been used in a number of aerosol models (Lurmann et al. 1997; Pilinis et al. 2000; Capaldo et al. 2000). An interpolation scheme is necessary for the implementation of this method in a multibox model because the aerosol distribution needs to be remapped onto a fixed size grid before it is exchanged between different boxes. Lurmann et al. (1997) used a spline interpolation scheme in their three-dimensional model.

In this paper, a one-dimensional Lagrangian trajectory model that employs the equilibrium, dynamic, and hybrid aerosol approaches is presented. The trajectory model is applied to an air pollution episode of 27–28 August 1987 in southern California during the Southern California Air Quality Study (SCAQS), and the results are compared to size-resolved aerosol measurements. Model predictions using different size resolutions are examined. To investigate the model response to emission changes, NO_x, VOCs, NH_3, and primary sulfate emissions are increased or decreased by 50% and $PM_{2.5}$ nitrate, PM_{10} sulfate, and total $PM_{2.5}$ predicted by 3 aerosol models are compared.

MODEL DESCRIPTION

The host trajectory model used in this work is the secondary organic aerosol model (SOAM), which contains interacting modules describing gas-phase chemistry, equilibrium partitioning of the condensable organic gases between the gas and aerosol phases, vertical dispersion of gas and aerosol species, emission of primary gaseous and aerosol pollutants, and dry deposition (Pandis et al. 1992).

Three aerosol modules were added to this one-dimensional framework to simulate condensation and evaporation of inorganic aerosol species. The equilibrium model employed in this work has been described by Capaldo et al. (2000). The amount of each species transferred between gas and aerosol phases is determined by bulk aerosol thermodynamics using ISORROPIA (Nenes et al. 1998) and is distributed over the aerosol size distribution by using a weighting factor based on the surface area of each size section (Pandis et al. 1993; Lurmann et al. 1997). The multicomponent aerosol dynamic model (MADM) developed by Pilinis et al. (2000) is the dynamic model used here. The mass transfer rates of aerosol species for each size section are determined by a fundamental flux equation (Seinfeld and Pandis 1998), and then constrained by limiting the acidity flux proportionally to the aerosol hydrogen ion concentration. Finally, the hybrid model developed by Capaldo et al. (2000) was incorporated into the trajectory model. In the original hybrid model, the dynamic calculation step followed the equilibrium step. In this study, the primary gas emissions are coupled with the gas-phase chemistry/diffusion operator. To assure that the coarse particles are exposed to some of the sulfuric acid vapor, we applied first the dynamic step and then the equilibrium step.

Mathematical descriptions of nucleation and coagulation have been added to all 3 approaches. The nucleation rate parametrization proposed by Russell et al. (1994) is employed in the model. The nucleation model used here assumes a linear sulfuric acid vapor concentration variation for the given timestep of the aerosol module based on the initially available sulfuric acid and also assigns all the nucleated mass to the first section of the distribution. The coagulation rate of the aerosol particles was modeled according to Seinfeld and Pandis (1998). A high-resolution distribution is used for the coagulation calculations by subdividing each section of the original distribution into 3 sections.

Since all 3 aerosol models internally use the moving sectional method, which allows the size sections to move in the size coordinate, a linear interpolation scheme was used to resize the aerosol particles between different computational cells by mapping the moving distribution onto a fixed size grid. While the spline interpolation method, which is currently used in

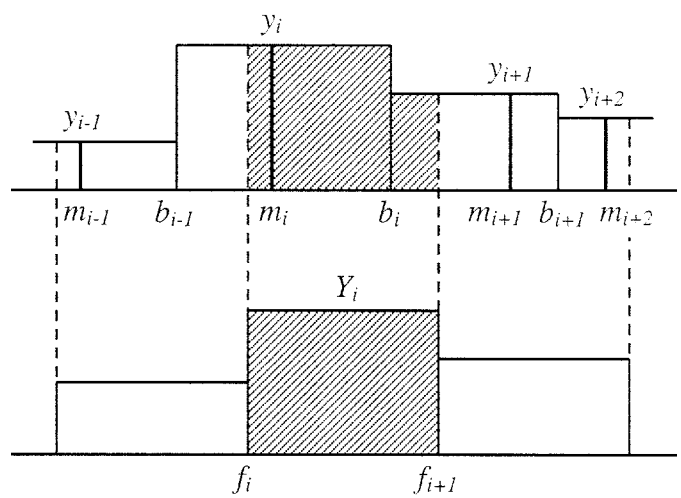

Figure 1. Schematic of the linear interpolation scheme; m_i is the logarithm of moving sectional diameters; b_i is the logarithm of boundaries of moving sections (the boundary diameter of a moving bin is calculated as the logarithmic mean of the 2 adjacent moving-bin center diameters), $b_i = (m_i + m_{i+1})/2$; f_i is the logarithm of fixed sectional diameters; y_i is the mass of aerosol species in a moving section; and Y_i is the mass of aerosol species in a fixed section, $Y_i = y_i(b_i - f_i)/(b_i - b_{i-1}) + y_{i+1}(f_{i+1} - b_i)/(b_{i+1} - b_i)$.

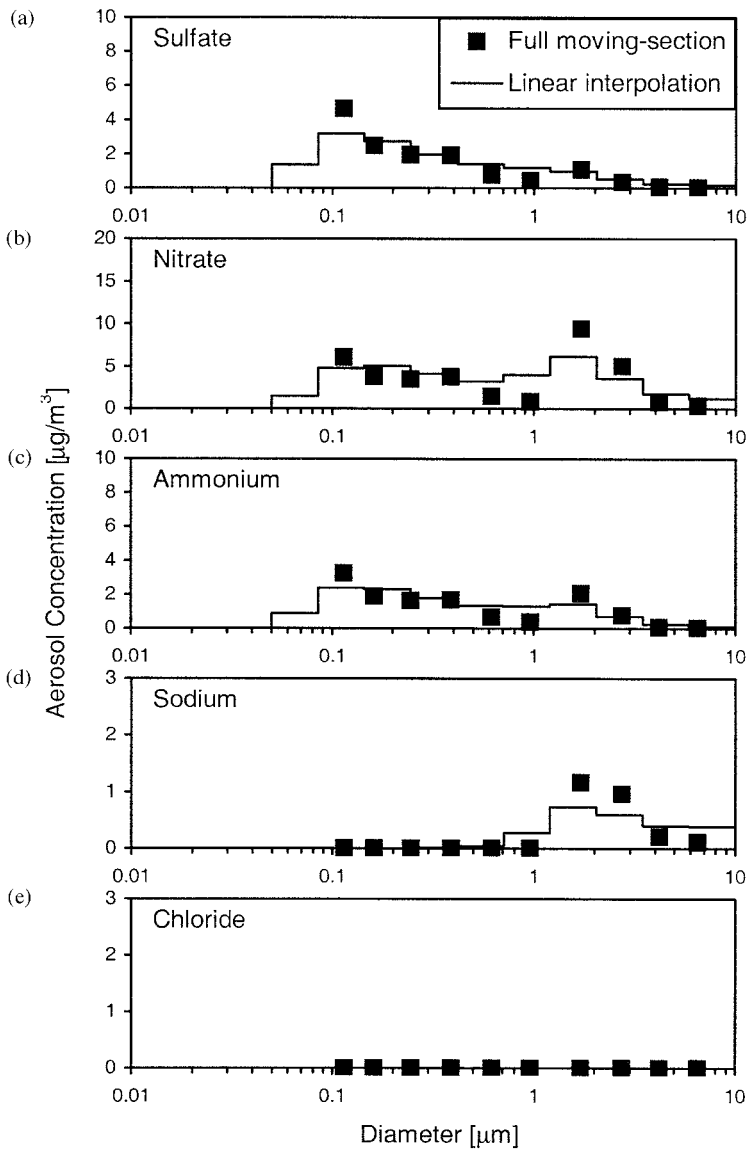

Figure 2. Aerosol size distributions predicted by a box model with and without the linear interpolation method. The initial distribution is the same as that of Pilinis et al. (2000).

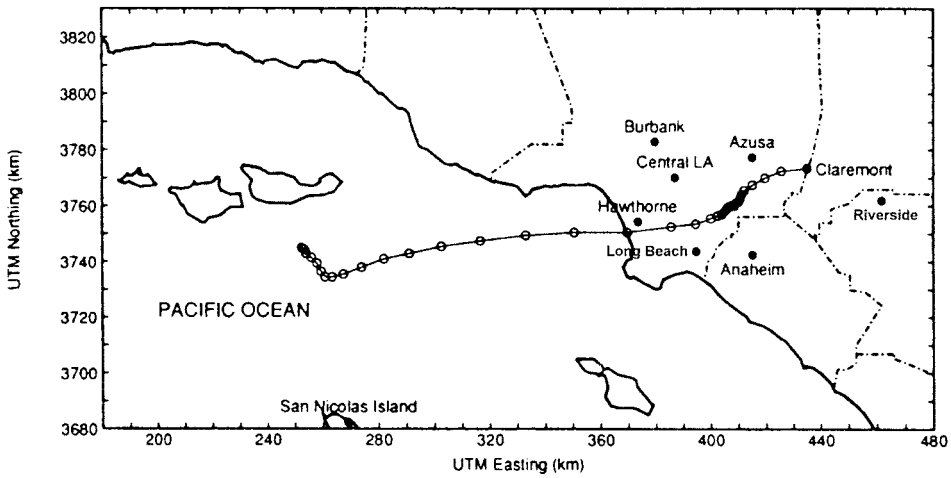

Figure 3. Trajectory of the air parcel arriving at Claremont on 28 August 1987 at 14:00 PST.

three-dimensional air quality models like UAM-AERO, works efficiently with equilibrium aerosol models (Lurmann et al., 1997), it can cause problems in the dynamic approach because the method cannot conserve some important aerosol properties, such as the charge balance established from the previous step of the aerosol model. The linear interpolation scheme used here distributes the aerosol mass of moving sections (in logarithmic size space) onto fixed size bins proportionally to the width of the moving section overlapping the corresponding fixed section (Figure 1). Mass is conserved while the number concentration is recalculated after interpolation at each time step based on the interpolated mass and center diameter of the fixed size section. Between interpolations, the particle diameters grow in exactly the same way as in a full moving-section method. In this scheme, acidity and other relevant properties calculated from interpolated aerosol mass stay within the upper and lower limits of the original moving sectional aerosol distribution. For example, consider the ratio of strong acids to bases (a measure of acidity) in section i, a_i, which is defined as

$$a_i = \frac{\sum_{j=anions} y_{i,j}}{\sum_{k=cations} y_{i,k}},$$

where $y_{i,j}$ is the mass of aerosol species j in the moving section i. After interpolation of the same ratio in the corresponding fixed size section, A_i is

$$A_i = \frac{\sum_{j=anions} Y_{i,j}}{\sum_{k=cations} Y_{i,k}} = \frac{C \sum_{j=anions} y_{i,j} + C' \sum_{j=anions} y_{i+1,j}}{C \sum_{k=cations} y_{i,k} + C' \sum_{k=cations} y_{i+1,k}},$$

$$C = \frac{b_i - f_i}{b_i - b_{i-1}}, \qquad C' = \frac{f_{i+1} - b_i}{b_{i+1} - b_i},$$

where $Y_{i,j}$ is the mass of aerosol species j in the fixed section i; b_i is the logarithm of the upper boundary of the moving section i; and f_i is the logarithm of the lower boundary of the fixed section i. Then it can be easily shown that

$$\min(a_i,\ a_{i+1}) \le A_i \le \max(a_i,\ a_{i+1}).$$

That is, the acidity of each interpolated section is always between the acidities of the 2 adjoining moving sections. Restriction of the ratio of the interpolated sections within those limits from the original moving sections is reasonable because acidities from the real aerosol size distribution would be smooth and continuous.

One problem with this type of linear interpolation method is numerical diffusion, which can be significant in the case of rapid growth (Jacobson 1997a). Methods that avoid this problem during the solution of condensation/evaporation (e.g., full moving-section method and moving-center method) usually introduce a similar error when particles are moved between adjacent computational cells in the atmospheric advection/dispersion calculations. Figure 2 shows the size distributions for the aerosol species after a 2-day simulation by a box model based on a scenario from Pilinis et al. (2000) with and without the linear interpolation method. The effects of numerical diffusion using 10 sections to cover $2^1/_2$ orders of magnitude are modest for sulfate and ammonium but more pronounced for nitrate. The distribution peaks are still at the right locations, and the method clearly outperforms approaches like the quasi-stationary method (Jacobson 1997b). The numerical diffusion error can be reduced further by use of additional sections.

The host model includes all 3 aerosol modules, and the user can select which approach is used for the simulation.

MODEL APPLICATION

The one-dimensional trajectory model described above was used to simulate the dynamics of the aerosol size/composition

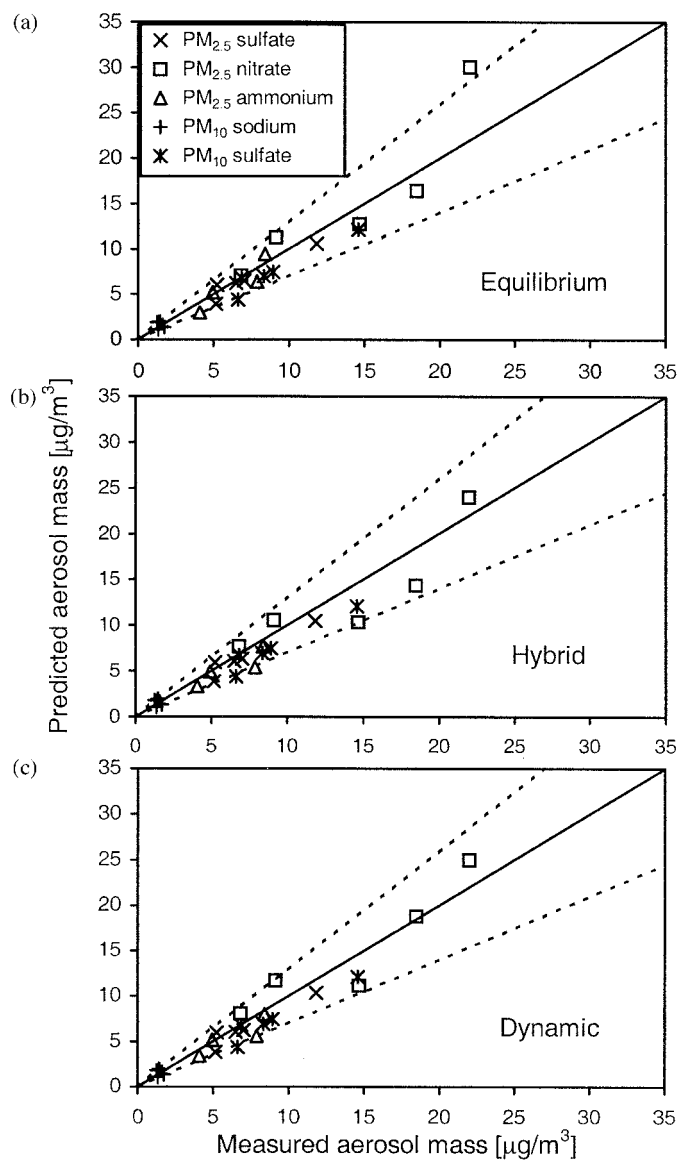

Figure 4. Predicted versus measured $PM_{2.5}$ and PM_{10} for aerosol species; solid lines are 1:1 correspondence lines and dotted lines represent an error of 30%.

distribution along 24 trajectories during the SCAQS episode of 27–28 August 1987. The air parcel trajectories start over the ocean at midnight (trajectories 1–16) or in the morning (trajectories 17–24) on August 27 and arrive at Claremont, CA at each hour of the day on August 28. Simulations were initiated 24 h before the beginning of the actual SCAQS episode in order to reduce the effect of the initial conditions on the results. Figure 3 shows one of the 24 trajectories that arrives at Claremont at 14:00 PST on 28 August 1987.

The one-dimensional modeling domain consists of 5 vertical cells with a height of 120 m each. The particle size distribution was divided into 8 size sections equally spaced in a logarithmic scale, where the lowest and largest size boundaries are 0.04 and 10 microns, respectively. In this grid, the boundary between the 6th and 7th sections corresponds to 2.5 microns. The threshold diameter used in the hybrid model is 0.625 microns, which corresponds to 4 sections simulated with the equilibrium and 4 with the dynamic approach. The time interval at which the equilibrium and dynamic step of the hybrid model take turns in the hybrid model is set to 2 min, which is large enough to achieve equilibrium in the fine mode but small enough to avoid errors from the decoupling of the fine and

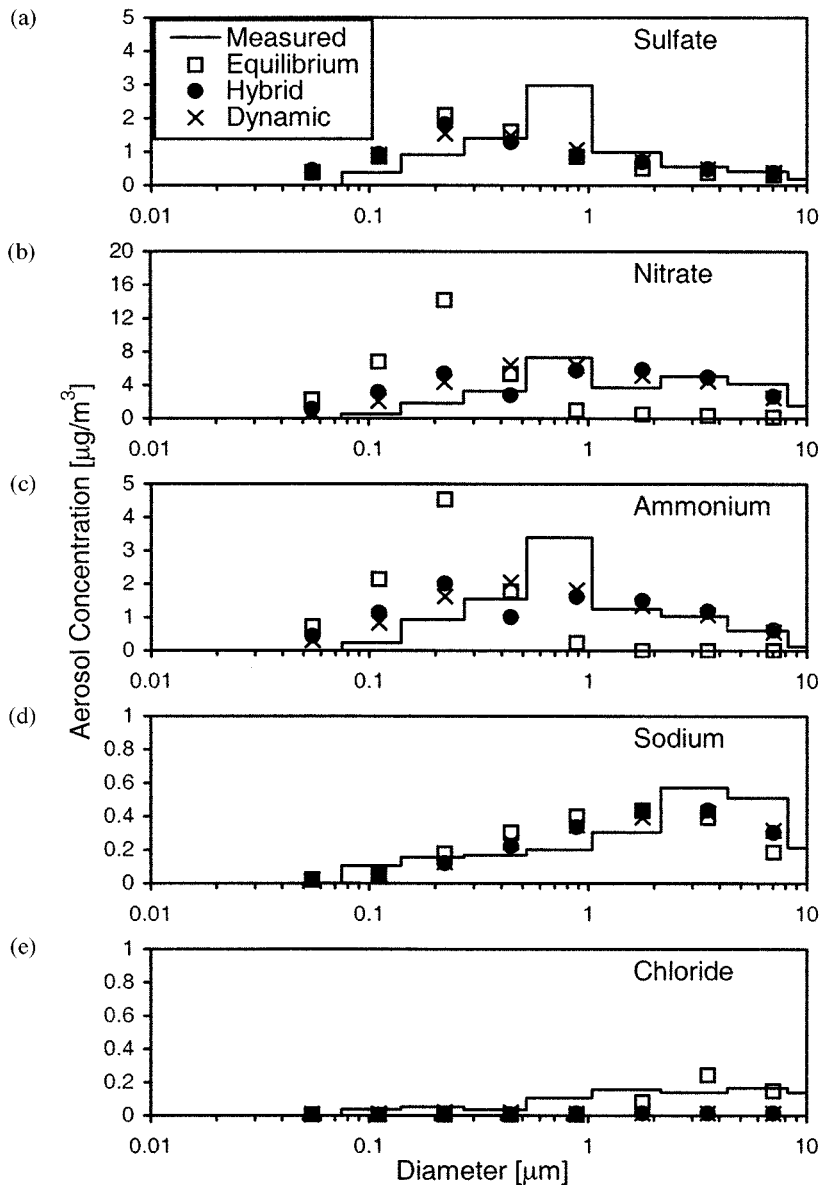

Figure 5. Predicted and measured aerosol distributions for the sampling period between 9:00 and 13:00 PST on 28 August 1987.

coarse particles. A timestep of 10 min was used for the aerosol operator.

For the evaluation of the model predictions, measurement data collected by a filter-based sampler (Fitz et al. 1989) and a 9 stage Berner impactor (John et al. 1990) were used for the $PM_{2.5}$ and PM_{10} aerosol species and aerosol size/composition distribution, respectively. Filter samples were collected during 5 sampling periods (0:00–5:00, 5:00–9:00, 9:00–13:00, 13:00–17:00, and 17:00–24:00 PST), and impactor samples were collected during 4 sampling periods (5:00–9:00, 9:00–13:00, 13:00–17:00, and 17:00–24:00 PST).

RESULTS AND DISCUSSION

The model predictions for the major aerosol species are compared to the available measurement data for the ground level in Figure 4. The filter measurements of PM_{10} nitrate and ammonium did not include the volatilized amount and so were not compared to the model predictions. All 3 methods reproduced all the 4–6 h averaged $PM_{2.5}$ and PM_{10} aerosol mass within 30% except for the $PM_{2.5}$ nitrate concentration predicted by the equilibrium model (one of the points had an error of 36%). The average relative errors are 19.4, 19.0, and 18.9% for the equilibrium, hybrid, and dynamic methods, respectively.

Predicted aerosol size distributions show a distinct difference among the 3 aerosol models (Figure 5). The equilibrium model fails to predict the ammonium nitrate coarse mode and overpredicts the fine mode for the nitrate and ammonium. The hybrid and dynamic models show better agreement with the observations. The overprediction of the fine mode and underprediction of the coarse mode in the equilibrium method are due to the "bulk aerosol treatment" and the weighting scheme, which assigns more condensates to smaller size sections. For the sulfate and ammonium distribution, all 3 models fail to predict the peak around 1 micron. One of the sources of this discrepancy is the aqueous-phase chemical reactions (Pandis and Seinfeld 1989), which have not been included in the model used in this study. When clouds or fogs exist in the modeling domain, the liquid water content significantly increases and aqueous-phase chemistry plays a major role. Another potential error source is the numerical diffusion caused by the interpolation step between the moving and fixed sections. Interpolating the sectionally-represented

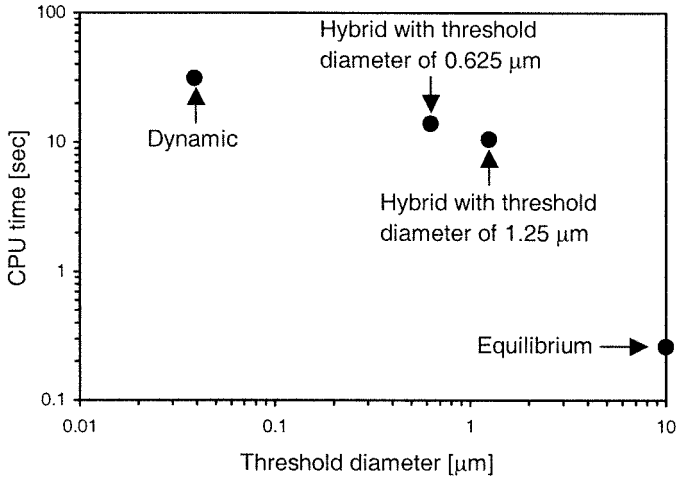

Figure 6. Average CPU time per simulation hour per computational cell versus equilibrium threshold diameter (on a 733 MHz Pentium III PC); an equilibrium threshold diameter equal to the lower boundary of the smallest size section corresponds to the dynamic method by definition. Similarly, the threshold diameter equal to the upper boundary of the largest size section corresponds to the equilibrium method.

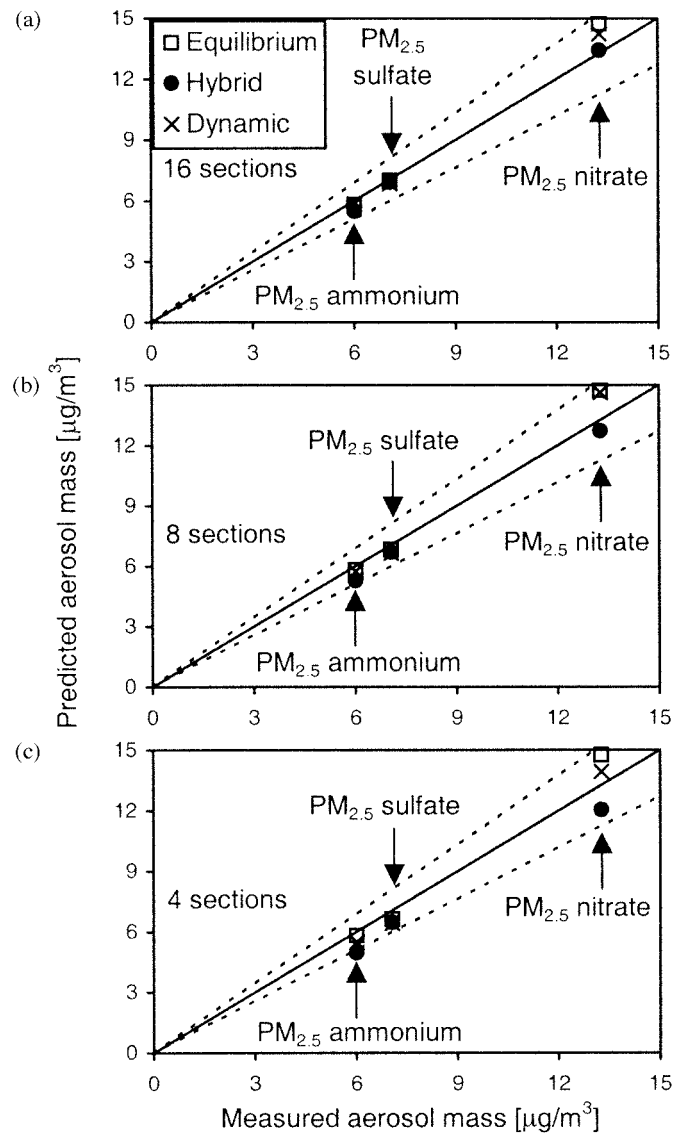

Figure 7. Predicted versus measured 24 h average $PM_{2.5}$ for aerosol species; the size resolutions used for the aerosol models are 4, 8, and 16 sections; solid lines are 1:1 correspondence lines and dotted lines represent an error of 15%.

size distribution tends to diffuse aerosol mass more or less along with diameter changes. Using a small number of sections in the sectional approximation required by the computational efficiency of three-dimensional models can make this error significant. The organic aerosol predictions are the same as in Pandis et al. (1992) and will not be discussed here.

Although the hybrid method is about 2–3 times faster than the dynamic method, it is still more demanding than the equilibrium method. Figure 6 shows the average CPU time per simulation hour per computational cell of the hybrid method according to the equilibrium threshold diameter.

SENSITIVITY TO SIZE RESOLUTION

In this work, 8 size sections have been used to facilitate the one to one comparison with the measurements. To examine the effects of the number of sections on the aerosol distribution, we have repeated the simulations using 4 and 16 sections. In the case of 16 aerosol size sections, the amount emitted in any of the 8 size bins of the emission data was evenly distributed to the corresponding 2 sections of the finer size distribution. In the case of the coarser aerosol distribution, the amount from each pair of the adjoining emission bins was added into the corresponding section of the 4 section aerosol distribution. Daily average $PM_{2.5}$ sulfate, nitrate, and ammonium predicted by all 3 aerosol models agreed with the measurement within 15% in all 3 resolutions except for the $PM_{2.5}$ ammonium by the hybrid model using 4 sections whose relative error is 18% (Figure 7). As the equilibrium model employs a weighting factor based on the surface area, the model is the least sensitive to size resolution. The hybrid model is more sensitive than the dynamic model because in the former the aerosol distribution is divided into 2 subdistributions: the equilibrium and dynamic sections. The subdistribution has fewer sections than the original one and so is more sensitive to the resolution. Although using 16 or 8 sections gives slightly better agreement with the observations, considering the computational efficiency and measurement error, 4 size sections can also be viewed as acceptable at least

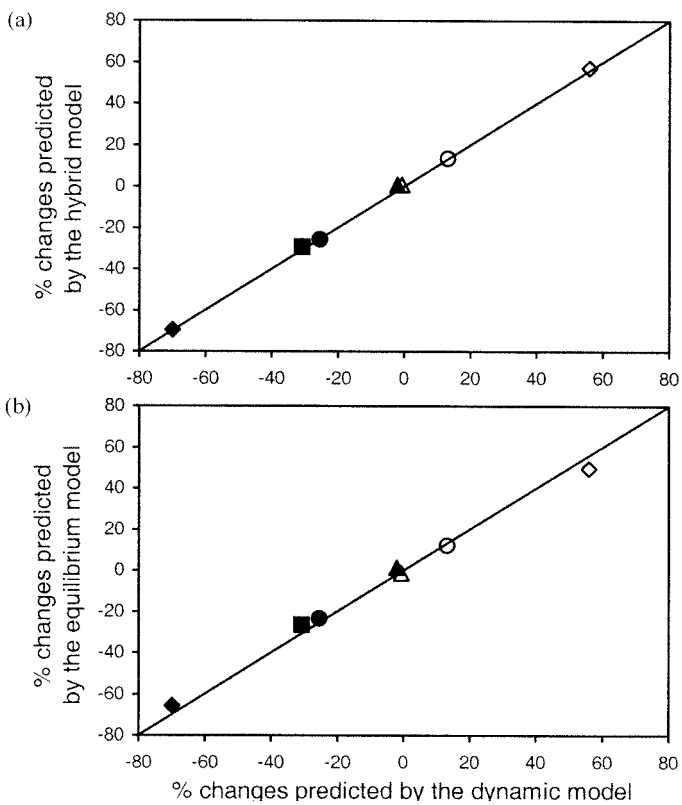

Figure 8. Percent changes of $PM_{2.5}$ nitrate concentrations predicted by the dynamic model versus the hybrid model and the dynamic model versus the equilibrium model for the selected emission changes: 50% decrease (■) in NO_x emissions; 50% decrease (●) and increase (○) in Volatile Organic Compound (VOC) emissions; 50% decrease (◆) and increase (◇) in NH_3 emissions; 50% decrease (▲) and increase (△) in sulfate emissions; solid lines are 1:1 correspondence lines.

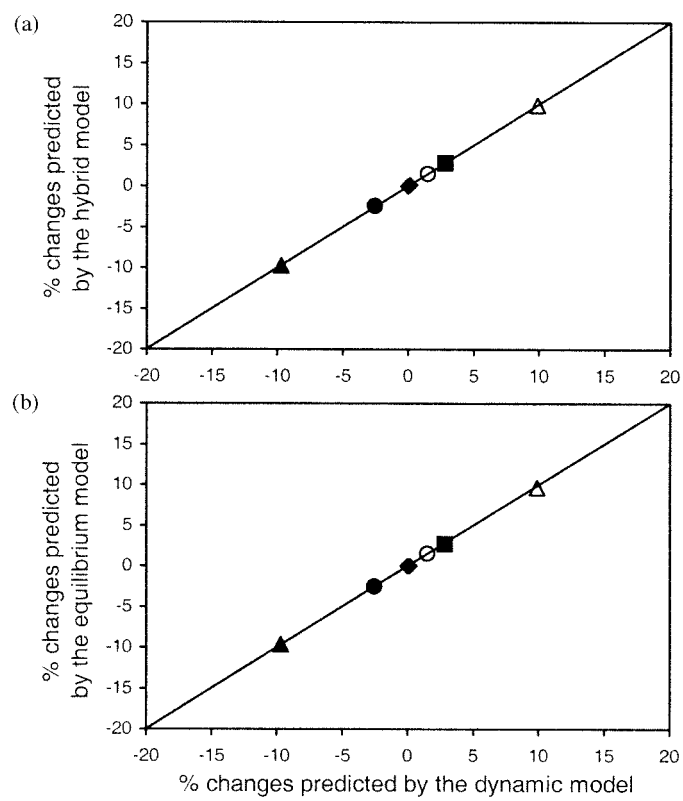

Figure 9. Percent changes of PM_{10} sulfate concentrations predicted by the dynamic model versus the hybrid model and the dynamic model versus the equilibrium model for the selected emission changes: 50% decrease (■) in NO_x emissions; 50% decrease (●) and increase (○) in VOC emissions; 50% decrease (◆) and increase (◇) in NH_3 emissions; 50% decrease (▲) and increase (△) in sulfate emissions; solid lines are 1:1 correspondence lines.

for this case in predicting daily average $PM_{2.5}$ concentrations. The higher resolution does not improve the equilibrium model predictions for the aerosol size distribution and makes only a small difference for the hybrid and dynamic models, while the lower resolution, as expected, hides the main features of the size distribution.

EFFICIENCY OF EMISSION CONTROLS

The predicted response to domain-wide emission changes was examined by changing NO_x, VOCs, NH_3, and primary sulfate emissions by 50%. The responses of predicted $PM_{2.5}$ nitrate, PM_{10} sulfate, and total $PM_{2.5}$ to emission changes for the trajectory arriving at 11:00 PST, where maximum $PM_{2.5}$ nitrate is predicted, are shown in Figures 8–10. A decrease in NO_x emissions for this case causes predicted $PM_{2.5}$ nitrate, and thus total $PM_{2.5}$, to decrease by about 30 and 10%, respectively. However, a reduction in NH_3 emissions results in about a 70 and 30% reduction of $PM_{2.5}$ nitrate and total $PM_{2.5}$, respectively. It has been reported that PM nitrate in this area is more sensitive to NH_3 than NO_x (Lurmann et al. 1997). Decreasing VOC emissions leads to decreases in both $PM_{2.5}$ nitrate and total $PM_{2.5}$ by reducing oxidant levels, which results in slower oxidation of NO_2 to HNO_3 (Lurmann et al. 1997). PM_{10} sulfate is little affected by changes in NO_x, VOC, and ammonia emissions, while changes in primary sulfate emissions directly affects the predicted concentration of PM_{10} sulfate. The effects of primary sulfate emission changes on $PM_{2.5}$ nitrate and total $PM_{2.5}$ are $< \pm 3\%$. The predicted responses by all 3 models are consistent for all cases. In general, the hybrid model response shows better agreement with the dynamic model than the equilibrium model does. However, the responses by both hybrid and equilibrium models agreed with those by the dynamic model within a few percent.

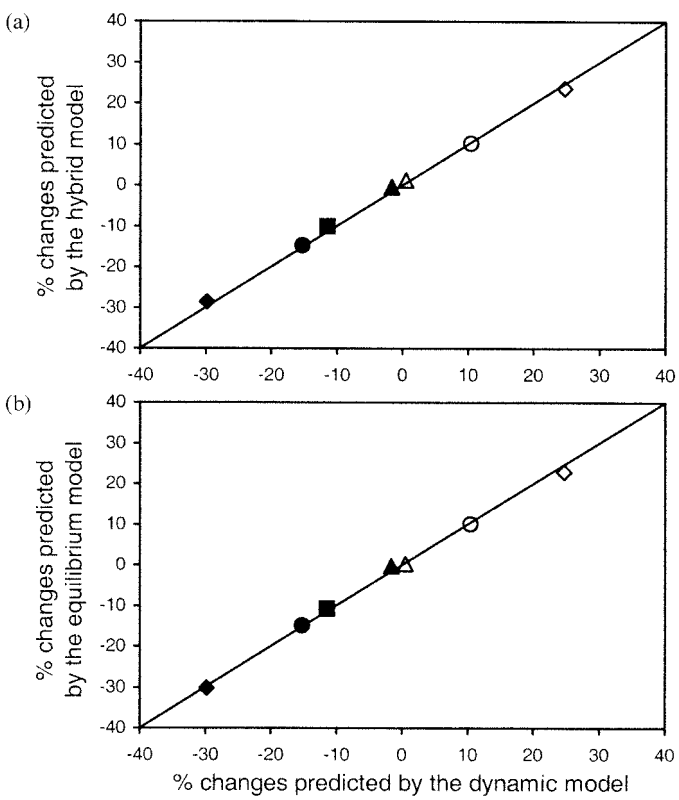

Figure 10. Percent changes of total $PM_{2.5}$ concentrations predicted by the dynamic model versus the hybrid model and the dynamic model versus the equilibrium model for the selected emission changes: 50% decrease (■) in NO_x emissions; 50% decrease (●) and increase (○) in VOC emissions; 50% decrease (◆) and increase (◇) in NH_3 emissions; 50% decrease (▲) and increase (△) in sulfate emissions; solid lines are 1:1 correspondence lines.

COMPARISON IN A THREE-DIMENSIONAL AIR QUALITY MODEL

The comprehensive air quality model with extensions (CAMx) was used as a host to compare the aerosol models in a three-dimensional framework (ENVIRON 2000). The equilibrium, hybrid, and dynamic aerosol modules were incorporated in CAMx to simulate the air pollution episode of 17 October 1995 in the Southern California air basin.

Performances of the 3 aerosol models for total $PM_{2.5}$, $PM_{2.5}$ nitrate, and $PM_{2.5}$ ammonium are compared in Figures 11–13,

Table 1
Twenty-four hour average concentrations averaged over 8 monitoring sites on 17 October 1995

	24 h average concentrations ($\mu g/m^3$)		
Species	Dynamic	Hybrid	Equilibrium
Total PM_{10}	44.6	44.3 (0.43)*	44.9 (0.62)
PM_{10} sulfate	1.7	1.7 (0.01)	1.7 (0.01)
PM_{10} nitrate	8.2	8.0 (0.31)	7.8 (0.51)
PM_{10} ammonium	2.6	2.5 (0.11)	2.8 (0.18)
Total $PM_{2.5}$	29.4	29.0 (0.53)	29.9 (0.75)
$PM_{2.5}$ sulfate	1.5	1.5 (0.01)	1.5 (0.01)
$PM_{2.5}$ nitrate	7.7	7.3 (0.38)	7.7 (0.45)
$PM_{2.5}$ ammonium	2.6	2.5 (0.13)	2.8 (0.20)
Total PM_1	23.7	23.0 (0.82)	24.4 (0.84)
PM_1 sulfate	1.3	1.3 (0.01)	1.3 (0.02)
PM_1 nitrate	7.4	6.8 (0.56)	7.7 (0.53)
PM_1 ammonium	2.5	2.4 (0.18)	2.7 (0.23)

*The numbers in parentheses are mean errors defined as follows:

$$\text{Mean error} = \frac{1}{N}\sum_{i=1}^{N}\left|c_i - c_i^d\right|,$$

where c_i is the 24 h average concentration at the ith monitoring site predicted by the hybrid or equilibrium model; c_i^d is the 24 h average concentration at the ith monitoring site predicted by the dynamic model; and N is the number of monitoring sites (Anaheim, Azusa, Burbank, Claremont, Hawthorne, Long Beach, Central LA, and Riverside).

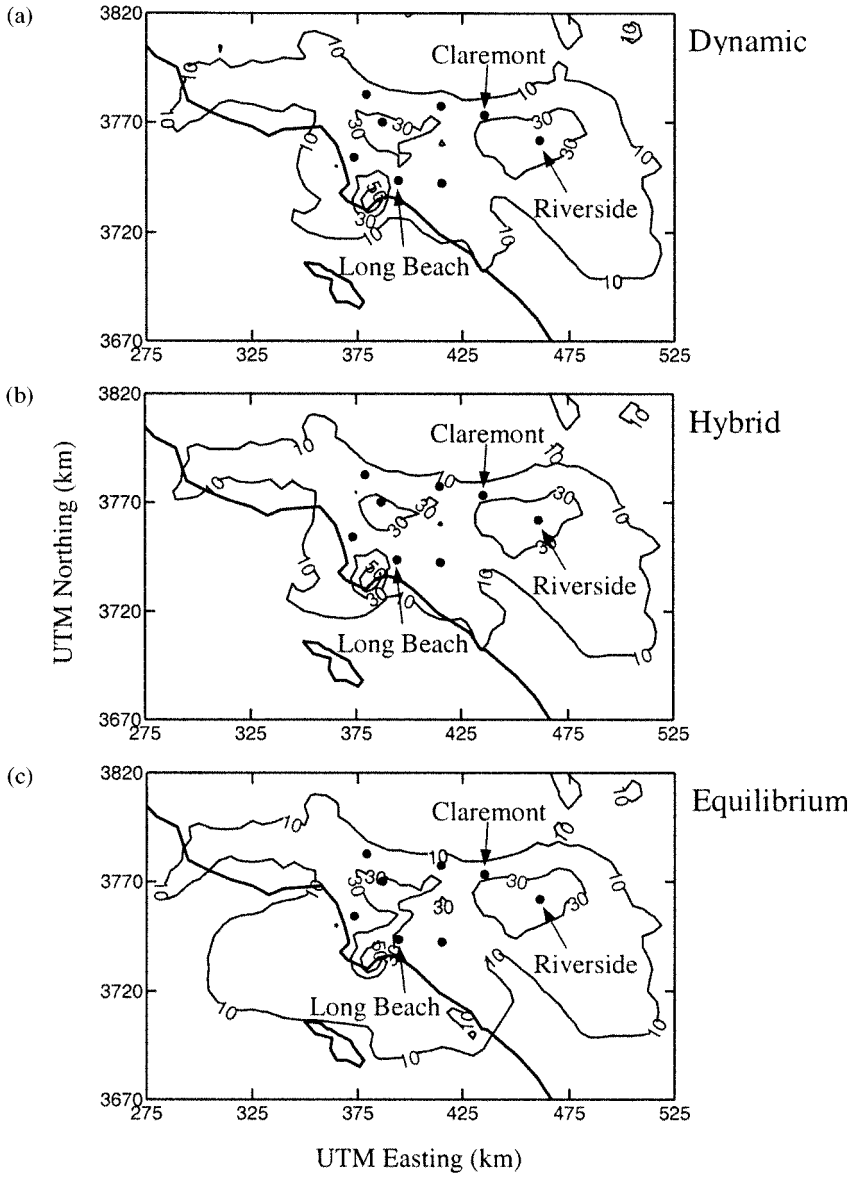

Figure 11. Predicted 24 h average total $PM_{2.5}$ concentrations ($\mu g/m^3$) by dynamic, hybrid, and equilibrium models on 17 October 1995 in the Southern California air basin.

respectively. The 24 h average total $PM_{2.5}$ and $PM_{2.5}$ ammonium reach their maximum near Long Beach on that day. The peak $PM_{2.5}$ nitrate concentration is predicted around the Riverside area. All 3 models show nearly identical spatial patterns over land. The equilibrium aerosol model tends to predict higher ammonium nitrate concentrations over the ocean because of the artificial mixing of the fine and coarse particles during the equilibrium calculation (Capaldo et al. 2000).

A statistical comparison of predictions by the 3 aerosol models is presented in Table 1. All 3 aerosol models are in good agreement. It should be noted that the mean errors for PM_1 (particulate matter with diameter $\leq$1 microns) species are higher than those for PM_{10} and $PM_{2.5}$ species. The overprediction in the equilibrium model is more significant for $PM_{2.5}$ and PM_1 species, as expected. The hybrid model tends to underpredict in the fine mode because the dynamic part (coarse mode) of the model interacts with the gas-phase species before the equilibrium part (fine mode) does. In general, the predictions by the hybrid model show slightly better agreement with those by the dynamic model.

CONCLUSIONS

In this work, 3 aerosol modules are incorporated into a one-dimensional Lagrangian trajectory model to compare their ability to predict the size/composition distribution as well as $PM_{2.5}$

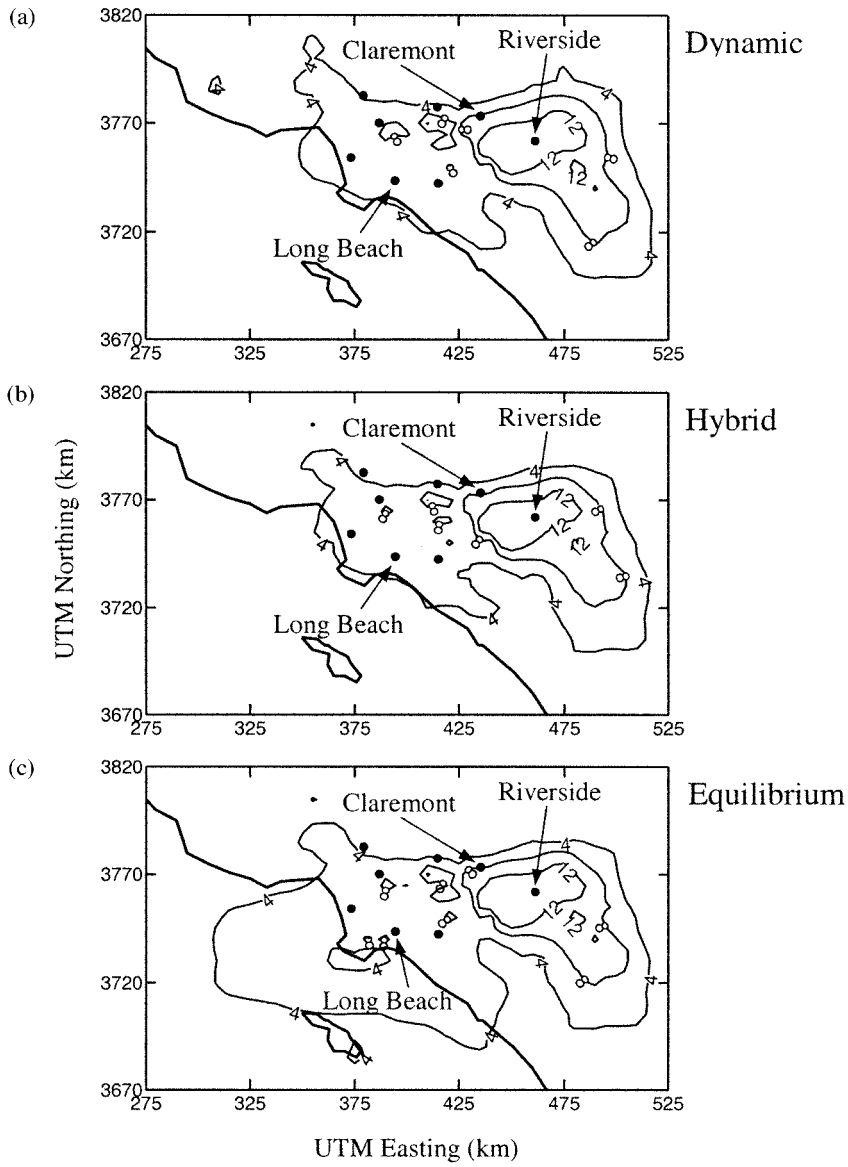

Figure 12. Predicted 24 h average $PM_{2.5}$ nitrate concentrations ($\mu g/m^3$) by dynamic, hybrid, and equilibrium models on 17 October 1995 in the Southern California air basin.

and PM_{10} concentrations of the major inorganic aerosol species. The model simulates gas and aerosol phases along the air parcel trajectories arriving at Claremont on 28 August 1987.

Although the predictions of $PM_{2.5}$ and PM_{10} aerosol species from all 3 aerosol models agreed reasonably well with the measurement data, the hybrid and dynamic models generally give better performance than the equilibrium model in predicting the aerosol size distribution. The comparison of aerosol models in a three-dimensional framework also shows consistent results with the one-dimensional model.

Computational efficiency is an important factor in the model applicability to a large-scale air quality model. The one-dimensional model used for the base case has 5 vertical cells, 8 aerosol size sections, and 25 organic and inorganic species interacting between gas and aerosol phases, which results in 1,000 differential equations. Although the computational efficiency of the hybrid model depends on the choice of the threshold diameter, the equilibrium model has a great advantage in its speed and stability.

To investigate model sensitivity to size resolution, the base case simulation was repeated with the resolution doubled or reduced by half. Even in the lowest resolution, 24 h average concentrations of $PM_{2.5}$ aerosol species predicted by all 3 models agreed with the observations in the measurement error range. The responses of $PM_{2.5}$ nitrate, PM_{10} sulfate, and total $PM_{2.5}$ to the changes in NO_x, VOCs, and primary sulfate emissions are compared to evaluate the model's response to emission changes. All 3 models show consistent predictions. The equilibrium

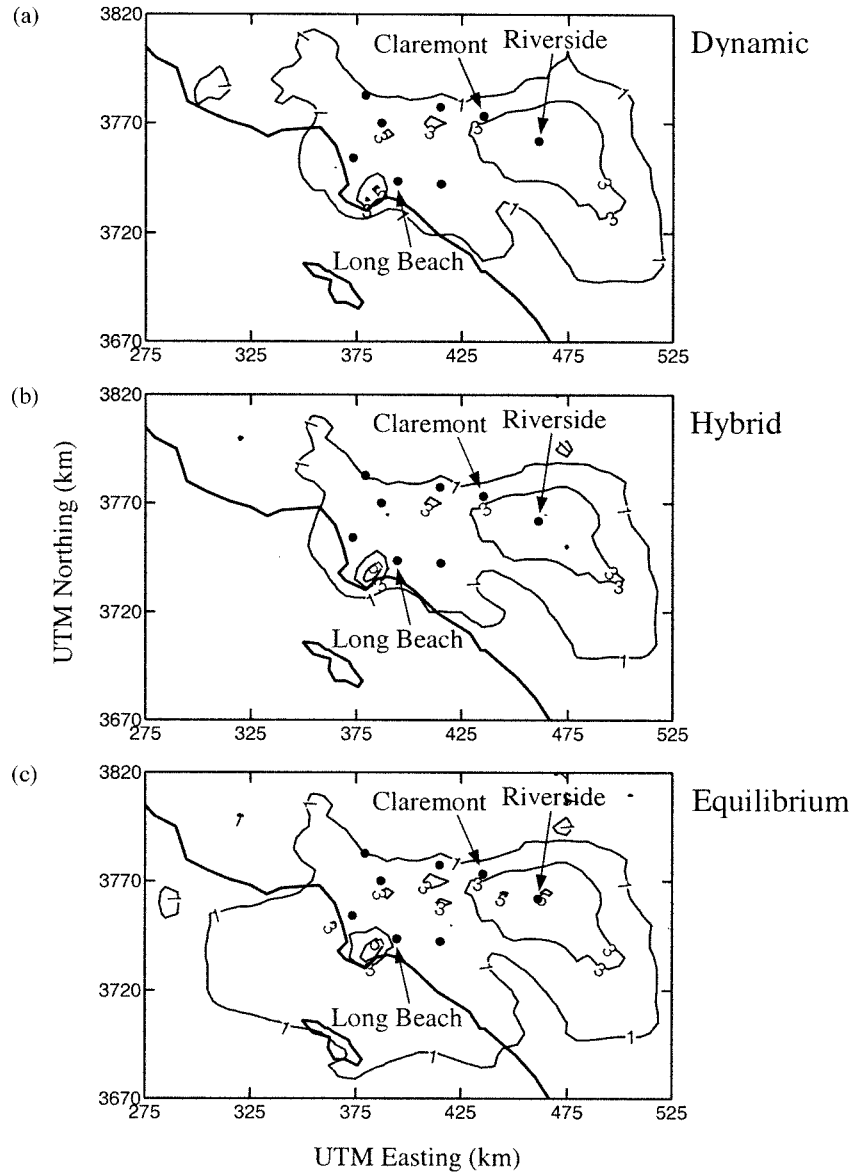

Figure 13. Predicted 24 h average $PM_{2.5}$ ammonium concentrations ($\mu g/m^3$) by dynamic, hybrid, and equilibrium models on 17 October 1995 in the Southern California air basin.

model is generally a little more sensitive to emission changes than the dynamic and hybrid models.

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