Technical Note:

Numerical instability in the Community Multiscale Air Quality model and its impacts on aerosol and ozone simulations

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Abstract

This paper reports a numerical instability problem in the widely used Community Multiscale Air Quality (CMAQ) model and discusses its impacts on ozone and particulate matter simulations. By adding 0.5 moles/sec of NO_x emissions to Middlesex County, CT, for example, CMAQ (2003 version) predicts up to 1 $\mu g/m^3$ change in $PM_{2.5}$ concentrations in the Ohio Valley and southern California in less then 48 hours. These regions are beyond the reach of normal transport processes in such a short time, and the remote and upwind responses are 100 times larger than responses near or downwind of the source area. More recently, progress has been made in reducing the numerical instability by correcting coding errors in the transport algorithm, adopting additional vertical wind adjustment to enhance mass conservation, and making numerous improvements in the ISSOROPIA aerosol thermodynamics module (2004 and 2005 CMAQ versions). These improvements, however, are not sufficient to reduce the instability to a reasonable level. The magnitude of peak instability in the 2005 version of CMAQ remains comparable to the normal responses from NO_x emissions of a middle-size power plant. This problem, although having a minor effect on the model performance to simulate total O_3 and PM concentrations, results in difficulties when the current version of CMAQ is used to address many important air quality issues including localized emission controls and source-receptor simulations.

Keywords: *CMAQ; Particulate matter (PM); Ozone (O_3); numerical instability; source-receptor; emission control*

1. Introduction

This paper reports a numerical instability problem in the widely used Community Multiscale Air Quality (CMAQ) model that has not been completely resolved. Air quality models such as CMAQ are valuable tools for simulating the spatial and temporal distributions of ozone (O_3) and particulate matter (PM) concentrations resulting from both local emissions and long-range transport in locations where measurements are not available. In addition, they allow an exploration of emission control strategies to mitigate

elevated concentrations of O_3 and PM, which can help states develop plans to attain the National Ambient Air Quality Standards (NAAQS). The EPA's CMAQ model (Byun and Ching, 1999) is being increasingly used by both the regulatory and scientific communities to investigate these issues. For instance, the recent Clean Air Interstate Rule (CAIR) study used an updated 2003 version of CMAQ to evaluate the effect of the rule in reducing PM concentrations and incidences of nonattainment of O_3 and PM National Ambient Air Quality Standards (NAAQS) in the eastern United States (US). We report here a numerical instability problem existing in recent versions of CMAQ that severely compromises the ability of CMAQ to address important air quality issues such as the ones above.

This paper begins with an explanation of this instability problem and shows modelers how to identify the problem. We then discuss the impact of the instability on CMAQ's ability to accurately simulate changes in PM and O_3 concentrations resulting from localized changes in emissions. A brief discussion of possible causes of the problem follows. We end with the current status of efforts made by the community towards reducing the numerical instability.

2. Methods

We use the CMAQ model to simulate the transport, chemical transformation, and deposition of O_3, PM, and their precursors for July 1996 over the continental United States. The meteorology and emission input data are the same as that used in Tong and Mauzerall (2005). The model domain covers the 48 contiguous US states and surrounding areas, with a horizontal grid resolution of 36×36 km^2. There are 12 vertical layers extending from the surface to approximately 15 km. Different from Tong and Mauzerall (2005) which used version 2002 of CMAQ to avoid numerical instabilities in simulated surface O_3 concentrations, here we examine the numerical problem using CMAQ version 2003 and later. The model is configured to include detailed implementation of horizontal and vertical advection based on the Piecewise Parabolic Method (PPM), turbulent diffusion based on K-theory, chemistry in the gas, liquid, and particulate phases using a modified version of the CBM-IV chemical mechanism (Gery et

al., 1994) (CB4-AQ-AE3), dry deposition, and cloud physics and chemistry (CLOUD-RADM).

We conduct three simulations, one with standard emissions and two perturbation simulations with small localized increases in the standard emission of nitrogen oxides (NOx = NO + NO_2). To examine the numerical instability, we subtract results of the standard simulation from each of the perturbation simulations and examine resulting changes in PM and O_3 concentrations across the continental United States.

3. Results and discussion

To demonstrate the numerical instability problem in CMAQ, Figure 1 shows the change in surface $PM_{2.5}$ concentrations resulting from the addition of 0.5 moles/sec NO_x emissions to Middlesex County, CT. The additional NOx emissions are distributed across the county in proportion to the baseline emissions. The responses in frame I of Fig. 1 are expected as the increased NO_x emissions generally lead to PM formation near the emission source and in downwind areas. The large responses of $PM_{2.5}$ concentrations in the Ohio Valley (frame II) and southern California (frame III) and surrounding areas, however, are not expected and do not appear to be realistic. The prevailing wind during this period is southwesterly in the northeast domain and these areas are far upwind of the emission sources. Frames II and III in Fig. 1 contain both increases and decreases in $PM_{2.5}$ concentrations. The grid cell of the largest increase in $PM_{2.5}$ concentrations is adjacent to that of the largest decrease. Variations in California represent the largest cross-domain responses, 100 times larger than those near or downwind of the sources. This region, taking into account the mean wind, is far beyond the reach of normal transport processes in such a short time.

The species contributing to the $PM_{2.5}$ instability vary with location. For the southern California area, 99.9% of the difference in $PM_{2.5}$ concentrations between the standard and perturbation simulations is due to aerosol nitrate and ammonium. Table 1 shows the major contributing species for the grid cells where the largest positive and negative responses occurred in Figure 1. For the cell with the largest negative response, aerosol nitrate contributes 78% of $\Delta PM_{2.5}$ (with 75% coming from the accumulation mode) and ammonium contributes the remaining 22% (with 21% coming from the accumulation

mode). For the cell with the largest positive response, virtually all of the increase is due to increased aerosol nitrate concentrations (with 96% from the accumulation mode). Over the Ohio Valley (not shown in Table 1), aerosol sulfate can contribute up to 0.25 $\mu g/m^3$ of the $PM_{2.5}$ change or approximately 30% of total mass concentration. For some other locations, organic aerosols of biogenic origin also contribute to the $PM_{2.5}$ instabilities, but at much lower magnitude (~0.01 $\mu g/m^3$ observed in this study).

To examine the extent of the numerical instability problem, we repeat the above simulations by increasing NO_x emissions from Georgia, a Southeast US location, by 10 moles/sec distributed across the entire state in proportion to baseline emissions. Similar numerical artifacts in the Ohio Valley and California are found, although the locations and magnitudes are slightly different from that in the Connecticut case.

To trace how the tiny increase in NO_x emissions results in a large response far upwind, we convert the 3-D concentration fields into a zonal sum along column lines so that we can examine the vertical and horizontal propagation of the numeral instability simultaneously. Figure 2 presents the zonal sum of aerosol nitrate concentration changes for the first eighteen hours during the Georgia simulation. The aerosol nitrate concentration artifacts occur in the upper layers of the western part of the US domain in less than three hours. The western instabilities are larger than the actual photochemical responses in the eastern part of the US domain in all layers. The instabilities grow with time and spread to lower altitudes while remaining large in the upper layers. Only a small portion of the instabilities can be observed at the surface through subsidence at locations such as California and the Ohio Valley. By the end of the 18 hour simulation, the instabilities have propagated to almost the full width of the top three layers, which extend from 650 to 200 mbar.

Speculation on factors contributing to the artifacts includes errors in the transport algorithm, mass conservation errors, and errors in the implementation of aerosol physics and chemistry in this version of CMAQ. A coding error was reported in the vertical and horizontal routines that implement the Piecewise Parabolic Method (PPM) for gas-dynamical simulations in the CMAQ 2003 version (Dr. Daewon Byun, personal communication). The error was caused by the fact that CMAQ assumes a uniform grid while the vertical layer spacing is non-uniform. The coding error in vertical PPM also

causes, in addition to a transport problem, a severe mass conservation problem (Hu et al., 2005). Hu et al. (2005) demonstrated that in the 2003 version of CMAQ the total mass of added tracer can be artificially increased five-fold within 40 hours. Therefore, any CMAQ run prior to correcting the coding error in vertical advection may be subject to severe mass conservation errors over complex terrain.

Incorporating the corrected advection module (in version 2004) and additional vertical wind adjustment (in version 2005) into CMAQ significantly reduces the numerical instability. These improvements, however, are not sufficient to satisfactorily eliminate the instabilities, particularly in the upper layers. The remaining instability is traced to ISORROPIA (Nenes et al., 1998), the thermodynamic module used in CMAQ to simulate aerosol formation from inorganic precursors and water. Bhave et al. (2004) found that at low relative humidity, infinitesimal changes in SO_4 or NH_x (the sum of NH_3 and NH_4) can shift NO_3 from the gas to aerosol phase, resulting in up to 10 $\mu g/m^3$ changes in aerosol nitrate concentrations. This explains why most instability is found in the upper layers where relative humidity (RH) values are low. The discontinuous changes in the partitioning of HNO_3/NO_3 between gas and aerosol phases will affect the concentrations of OH and NO_2, via $HNO_3 \rightarrow OH + NO_2$. This in turn effects simulated O_3 concentrations as demonstrated by the response in O_3 concentrations. Fig. 3 shows the difference in O_3 concentrations between the same baseline and perturbation runs as in Fig 2. Similar to aerosols, O_3 concentrations also display unexpected responses in the western part of the domain. O_3 instabilities roughly co-locate with those of the aerosol nitrate instability. Disabling the aerosol thermodynamics in the CMAQ simulations can completely eliminate the artifacts in O_3 responses, suggesting that the O_3 instability is also linked to the ISSOROPIA module.

The latest version of CMAQ (released on September 24, 2005 by the US EPA) has reflected substantial efforts by the community to address the above problems. These include a corrected advection module, adoption of the vertical wind adjustment to improve mass conservation, and an updated ISORROPIA module. Repeating the above simulations with this version reveals the instabilities have been reduced by 100 times in most of the domain. The peak instability, however, remains notable with 0.1 $\mu g/m^3$ at the surface and up to 1 $\mu g/m^3$ PM in the upper layers upwind of the emission source. This

magnitude is comparable to the normal responses from NO_x emissions of a middle-size power plant (say 10 thousand tons NO_x per year).

The numerical instability we report here makes the use of current versions of CMAQ inadequate for applications that examine the effect of small changes in emissions on ambient concentrations of PM. However, it does not appear to have a significant effect on simulations of the total concentrations of PM and O_3. In fact, model evaluation studies show that the model is generally doing a reasonably good job simulating the temporal and spatial distributions of surface O_3 (e.g., Tong and Mauzerall, in press, 2005) and PM (e.g., Eder and Yu, accepted, 2005) over the continental United States. The traditional method of model evaluation in which simulated and measured concentrations of chemical species are compared, however, is not sufficient to either identify or probe model problems such as the numerical instability we report here.

The 2003 version of CMAQ and later versions that have not adopted necessary corrections in the aerosol chemistry and transport modules have been widely used in a number of scientific and regulatory applications. Some of these applications may suffer severely from the numerical instabilities in PM concentrations and to a lesser degree in O_3 concentrations and should be repeated with an updated version of the model. In addition, further work is needed to reduce the numerical artifacts that continue to result from small perturbations to NO_x emissions.

4. Conclusion

We report a numerical instability problem in the widely used Community Multiscale Air Quality (CMAQ) model. When we add 0.5 moles/sec NOx emissions to Middlesex County, CT, CMAQ (2003 version) predicts up to 1 $\mu g/m^3$ change in $PM_{2.5}$ concentrations in the Ohio Valley and southern California in less than 48 hours. These regions are beyond the reach of normal transport processes in such a short time, and the remote and upwind responses are 100 times larger than these near or downwind of the source area. Recent releases of CMAQ (2004 and 2005 versions) have made progress in reducing the numerical instability by correcting coding errors in the transport algorithm, adopting additional vertical wind adjustments to enhance mass conservation, and making numerous improvements in the ISSOROPIA aerosol thermodynamics module. These

improvements, however, are not sufficient to satisfactorily eliminate the instabilities. The peak instability in the latest CMAQ version (2005) remains large with 0.1 $\mu g/m^3$ at the surface and up to 1 $\mu g/m^3$ in the upper troposphere resulting from the 0.5 moles/sec increase in NOx emissions, a magnitude comparable to the normal responses from NO_x emissions of a middle-size power plant. This problem, if unresolved, will hinder the ability of CMAQ to address many important air quality issues of scientific and regulatory importance including evaluations of source-receptor relationships and the effects of regional cap-and-trade programs.

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Table and Figures

Table 1. Relative contribution of aerosol species to the $PM_{2.5}$ instabilities in the Connecticut simulations. Both the largest increase and decrease in $PM_{2.5}$ concentrations at the surface are found in California.

Fig. 1. Change in surface $PM_{2.5}$ concentrations resulting from the 0.5 moles/sec NO_x emissions added to Middlesex County, CT.

Fig. 2. Horizontal and vertical propagation of the numerical instability in aerosol nitrate concentrations.

Fig. 3. Same as in Fig.2, but for O_3 concentrations.

Table 1. Relative contribution of aerosol species to the $PM_{2.5}$ instabilities in the Connecticut simulations. Both the largest increase and decrease in $PM_{2.5}$ concentrations at the surface are found in California.

Aerosol Species[a]	Largest negative response (23,34) [b] $\Delta PM_{2.5}$ = -1.186 μg/m^3		Largest positive response (19,32)[b] $\Delta PM_{2.5}$ = 0.345 μg/m^3	
	ΔConc (μg/m^3)	ΔPercentage	ΔConc (μg/m^3)	ΔPercentage
ΔANO3I	-0.039	3.3%	0.017	4.9%
ΔANO3J	-0.885	74.6%	0.333	96.5%
ΔANH4I	-0.011	0.9%	0.0	0%
ΔANH4J	-0.251	21.2%	-0.005	-1.4%

a. The definition of aerosol species is as follows:
ANO3I, Aitken mode aerosol nitrate;
ANO3J, Accumulation mode aerosol nitrate;
ANH4I, Aitken mode aerosol ammonium;
ANH4J, Accumulation mode aerosol ammonium.

b. Numbers in parenthesis indicate grid box at surface of model domain. Both grid cells are located in southern California.

Fig. 1. Change in surface $PM_{2.5}$ concentrations resulting from 0.5 moles/sec NO_x emissions added to Middlesex County, CT.

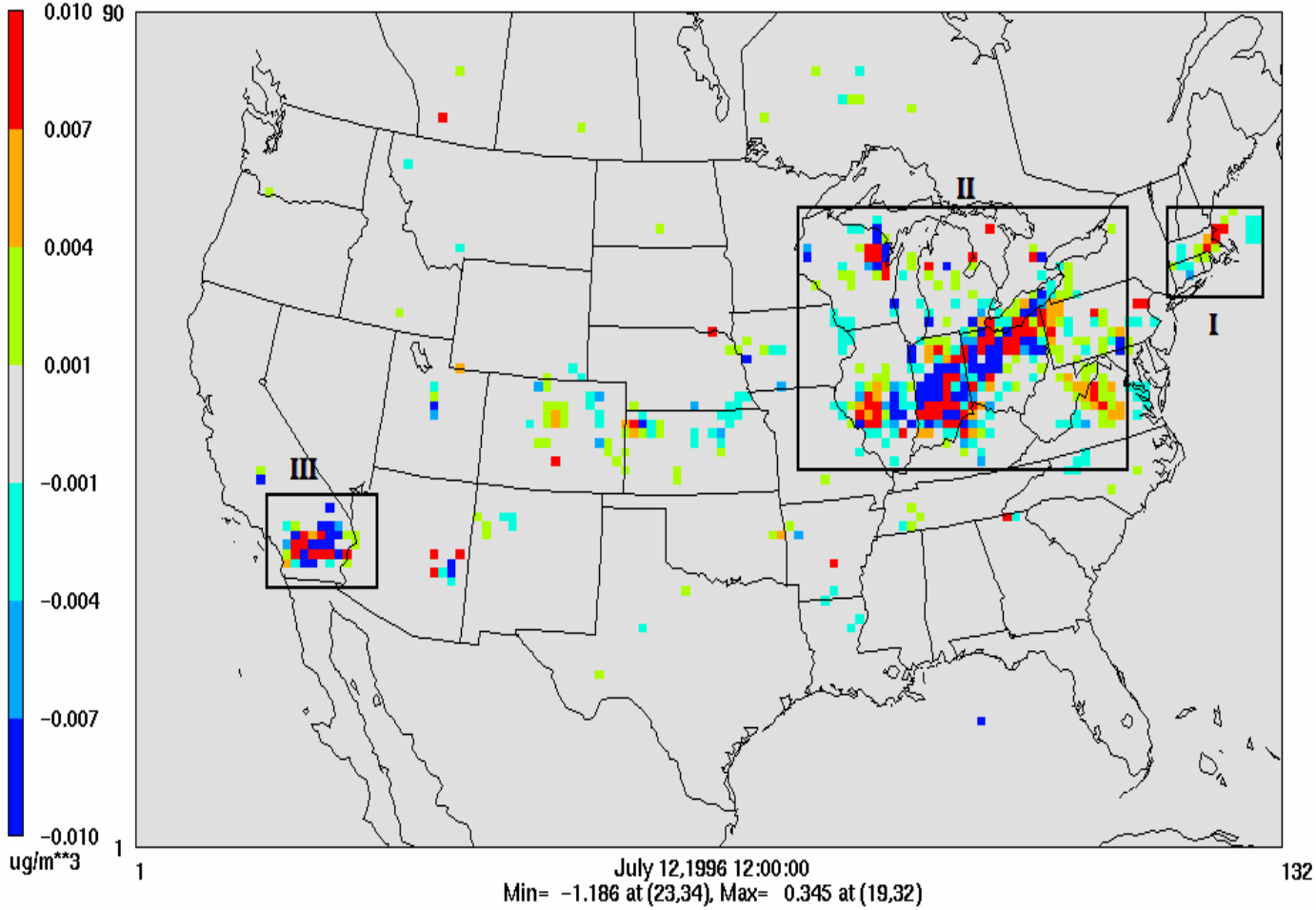

Fig. 2 Horizontal and vertical distribution of aerosol nitrate concentrations resulting from the addition of 10 moles/sec NOx emissions in Georgia, distributed across the state in proportion to the baseline emissions. The numerical artifact can be seen propagating over the 18 hour simulation. X marks the location of emissions of the additional NO_x. Grid box numbers on the horizontal axis can be mapped to spatial location by comparing with Figure 1. Vertical layers extend from the surface to approximately 15 km.

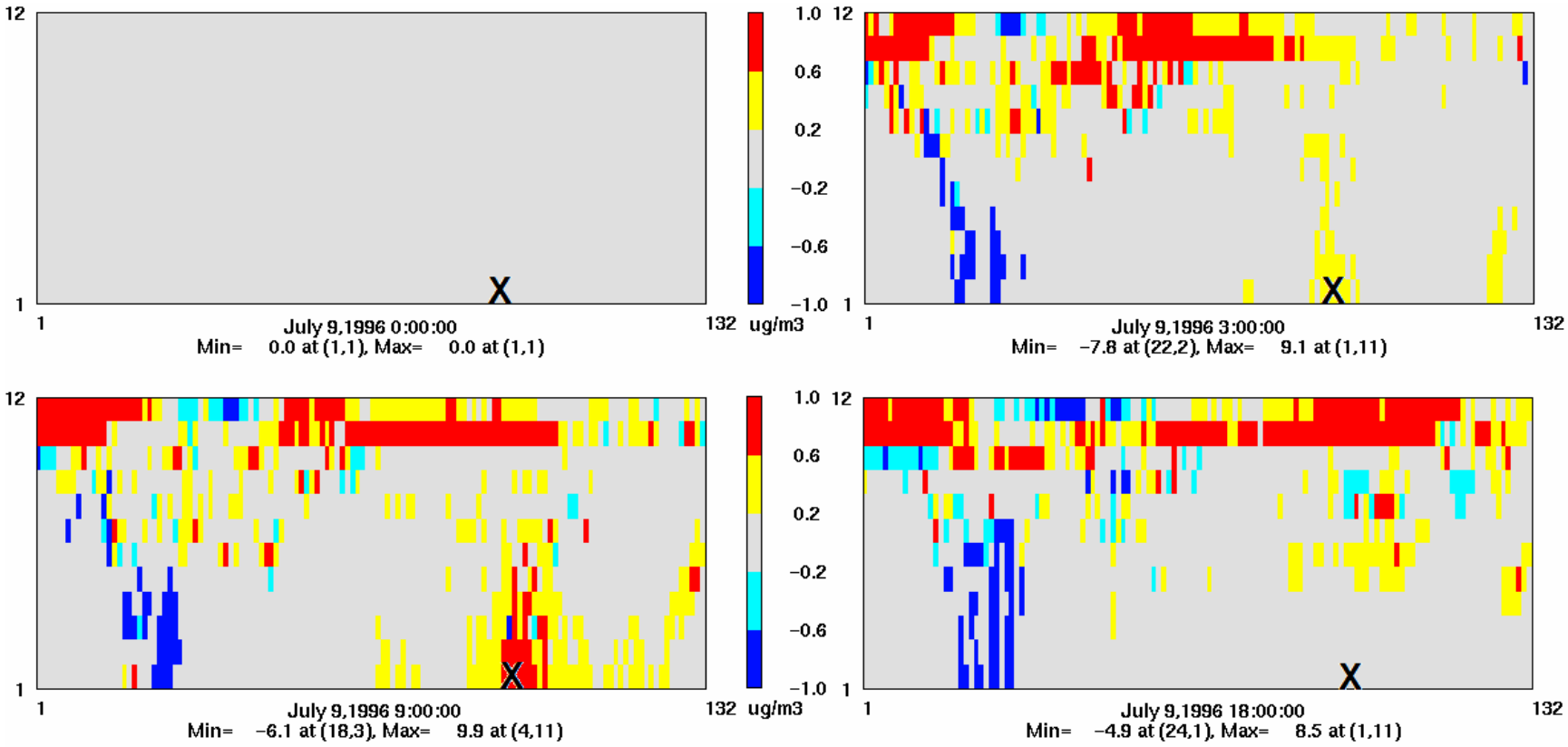

Fig. 3. Same as in Fig.2, but for O_3 concentrations.