CMAQ AEROSOL MODULE DEVELOPMENT: RECENT ENHANCEMENTS AND FUTURE PLANS

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1. INTRODUCTION

The Community Multiscale Air Quality (CMAQ) model has been used extensively in recent years to simulate ambient fine particle concentrations for both regulatory and research-grade applications. To keep pace with advancements in atmospheric aerosol research and to respond to various needs of the growing CMAQ user community, the aerosol module must be updated frequently. The aerosol module incorporated into the first public release of CMAQ in June 1998 is described in detail by Binkowski and Roselle (2003). In June 2002, a new aerosol module, `aero3`, was released with improved treatments of secondary organic aerosol (SOA) formation, sulfate nucleation, inorganic equilibrium partitioning, and heterogeneous nitrate formation (Schere, 2002). In September 2003, Version 4.3 of CMAQ was released with updates to the SOA and heterogeneous nitrate treatments (Pleim et al. 2003). The purpose of this paper is to describe aerosol module enhancements in CMAQ Version 4.4, released to the public in Fall 2004. The updates discussed herein are intended to enhance computational efficiency, improve numerical stability, streamline the code structure, and strengthen the in-line code documentation. In addition, aerosol module improvements anticipated to be in the 2005 CMAQ release are discussed briefly.

2. RECENT AEROSOL MODULE DEVELOPMENTS

2.1 Computational Efficiency

The CMAQ model runtime is affected by numerous factors, including the number and speed of computer processors, the number of grid cells in the domain, and the science modules selected. Computational-efficiency results presented in this section are obtained using a single Intel Xeon 3.06 GHz processor. Aerosol processes and aqueous chemistry are activated, and

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the Carbon Bond IV gas-phase chemical mechanism is used with the Euler Backward Iterative numerical solver (i.e., `cb4_ae3_aq` mechanism). The domain consists of 178×124 grid cells that span the continental United States with 32 km horizontal spacing and 21 vertical layers. Simulations begin with clean initial conditions on July 1, 1999, and the computational efficiency results reported below are based on Day 5 of the simulation.

The relative computer processor requirements of various science modules in CMAQ4.3 are illustrated in the left half of Figure 1. Aerosol module calculations (subroutine `AERO`) consume 44% of the total model runtime. Of this fraction, 47% is used to calculate coagulation coefficients (subroutines `INTERCOAG_GH` and `INTRACOAG_GH`), 31% is consumed by the ISORROPIA inorganic partitioning calculations (subroutine `EQL3`), and 13% is spent on SOA partitioning (subroutine `ORGAER3`).

The coagulation and SOA calculations have been modified in CMAQ4.4 to increase computational efficiency without compromising the overall model results. A new subroutine, `GETCOAGS`, has been added to calculate the coagulation coefficients in a more efficient manner than the Gauss-Hermite numerical quadrature algorithm used previously. In `GETCOAGS`, coagulation coefficients for particle number and aerosol third moment are calculated from analytical expressions reported by Binkowski and Shankar (1995). The second moment coagulation coefficients are calculated using analytical expressions for the near-continuum particle-size regime and correction factors for the free-molecular regime, similar to the approach outlined in Appendix H of the report by Whitby et al. (1991). The free-molecular correction factors are calculated offline using Gauss-Hermite numerical quadrature and retrieved from a lookup table in the `GETCOAGS` subroutine. The coagulation coefficients obtained from `GETCOAGS` differ from those of the CMAQ4.3 method on the order of one percent. These differences translate to an insignificant effect on the aerosol species concentrations when `GETCOAGS` is implemented in our CMAQ simulations. Users may evaluate the effect of the new coagulation routine on their respective model applications by toggling the value of `FASTCOAG_FLAG` in subroutine `AEROPROC`. The default value of this flag is `.TRUE.` in CMAQ4.4, to utilize the `GETCOAGS` subroutine. If
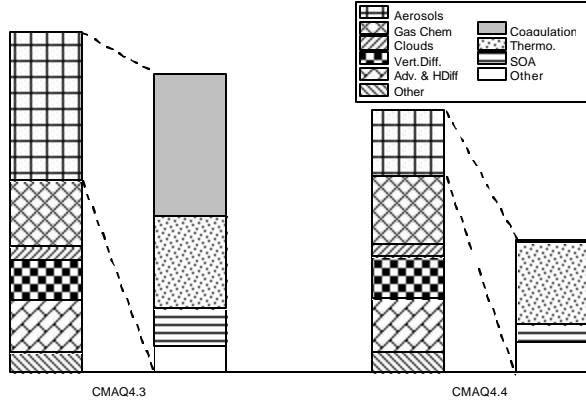
Figure 1. Relative amounts of computer processor time consumed by science modules and aerosol sub-modules in Versions 4.3 and 4.4 of CMAQ. Aerosol sub-module runtimes are magnified by a factor of two in the 2nd and 4th stacked bars.

changed to `.FALSE.`, the Gauss-Hermite algorithms will be employed in a manner identical to CMAQ4.3 and prior versions.

In the SOA partitioning algorithm in CMAQ, the most computationally demanding step involves solving a system of quadratic equations:

$$C_{aer,i} = C_{tot,i} - C_{sat,i}^* \frac{C_{aer,i}/m_i}{\sum_{j=1}^{n}(C_{aer,j}/m_j) + C_{pri}/m_{pri}} \quad (1)$$

where $C_{aer,i}$ is the aerosol-phase mass concentration of species i [$\mu g\ m^{-3}$], $C_{tot,i}$ is the total concentration of species i in both phases [$\mu g\ m^{-3}$], $C^*_{sat,i}$ is the temperature-dependent saturation vapor concentration of species i [$\mu g\ m^{-3}$], m_i is the molar mass of species i [g mol^{-1}], C_{pri} is the concentration of primary organic aerosol [$\mu g\ m^{-3}$], m_{pri} is the average molar mass of the primary organic aerosol [g mol^{-1}], and n is the number of semivolatile species that partition between the gas and aerosol phases. Note that the C_{aer} term appears on both sides of Equation 1 and cannot be solved analytically. Hence, this system of equations is solved iteratively in subroutine `NEWT` using a globally-convergent Newton-Raphson method (Schell et al., 2001).

As discussed by Pun et al. (2003), the quadratic system of equations can be reduced to a set of decoupled algebraic equations if one assumes that the total molar concentration of organic aerosol, n_{tot}, is constant during each model time step, i.e.,

$$n_{tot}(t) \approx \sum_{j=1}^{n}(C_{aer,j}(t_0)/m_j) + C_{pri}(t)/m_{pri} \quad (2)$$

where t_0 and t represent the beginning and end of the model time step, respectively. In CMAQ, the $C_{pri,i}(t)$ term is known a priori because primary emissions are accounted for before SOA partitioning as a result of operator splitting. Combining Equations 1 and 2 yields the following approximate analytic solution for $C_{aer,i}(t)$ after some algebraic manipulation:

$$C_{aer,i}(t) \approx C_{tot,i}(t) \frac{n_{tot}(t)}{n_{tot}(t) + C_{sat,i}^*(t)/m_i} \quad (3)$$

To gauge the effect of this approximation, model simulations similar to those outlined in Section 2.1 were conducted using Equation 3 instead of the iterative Newton-Raphson solution. A notable improvement in computational efficiency was realized, but model results were affected significantly. By assuming that n_{tot} is constant over each model time step (i.e., Equation 2), SOA concentrations decrease artificially in cells where the air is cooling rapidly and increase in cells where the air is warming rapidly. One effect of the approximation is an increase in afternoon SOA concentrations and a decrease in early-morning SOA concentrations over the southeastern U.S. by ~0.5 $\mu g\ m^{-3}$. To avoid introducing these errors, the constant n_{tot} approximation has not been incorporated in CMAQ4.4. Instead, the solution to Equation 3 has been adopted as the initial guess for the Newton-Raphson iteration. The modified initial guess substantially reduces the number of iterations required for convergence, relative to CMAQ4.3 wherein the initial guess is set to an equal distribution of each species between the gas and aerosol phases.

Effects of the aforementioned computational efficiency improvements on CMAQ runtime are displayed in Figure 1. Aerosol module calculations are a factor of 2.3 faster in CMAQ4.4 than in CMAQ4.3. Adoption of the `GETCOAGS` subroutine is responsible for 88% of this runtime reduction and the remaining 12% is due to the improved initial guess for the Newton-Raphson iterative solution. As shown in Figure 1, CMAQ4.4 aerosol module calculations contribute 25% of the total model runtime, comparable to the resource requirements of gas-phase chemistry (26%) and the sum of advection and horizontal diffusion (20%). Within the CMAQ4.4 aerosol module, 62% of the processing time is attributed to ISORROPIA inorganic equilibrium calculations, 14% to SOA calculations, 21% to miscellaneous aerosol calculations (e.g., solving the modal dynamics equations, mode merging, etc.), and 1.7% to coagulation calculations. Thus, future efforts to increase the CMAQ aerosol module efficiency should be directed at the inorganic equilibrium calculations.

2.2 Numerical Stability

During developmental testing of CMAQ, small numerical perturbations caused by the use of different FORTRAN compilers were found to produce large (~10 $\mu g\ m^{-3}$), transient impacts on aerosol-phase nitrate concentrations over the arid southwestern U.S. These effects have been attributed to numerical instabilities in the ISORROPIA thermodynamics module. To examine these instabilities, ISORROPIA was run independently with fixed input values of temperature, relative humidity (RH), total sulfate ($TSO_4 = H_2SO_{4(g)} + SO_4$), and total nitrate ($TNO_3 = HNO_{3(g)} + NO_3$), while varying the total ammonia ($NH_x = NH_{3(g)} + NH_4$) input. One example of numerical instabilities in ISORROPIA is illustrated in Figure 2a, where the aerosol-phase NO_3 concentrations

are plotted as a function of the molar ratio of NH_X/TSO_4. Under hot and dry conditions, nitrate should remain largely in the gas phase based on Gibbs free energy considerations. However, when the NH_X/TSO_4 ratio falls between 13.3 and 15.1 (i.e., $NH_X = 2.4 - 2.8$ μg m^{-3}) or above 21.5 (i.e., $NH_X > 4.0$ μg m^{-3}), the ISORROPIA module in CMAQ4.3 yields substantial quantities of aerosol-phase NO_3 (see solid line in Figure 2a). Through correspondence with the developer of ISORROPIA, Dr. Athanasios Nenes, the causes of these anomalous results were identified. The discontinuous results at intermediate NH_X/TSO_4 values are due to a numerical error in the `EX10` function subprogram. The production of aerosol NO_3 at high NH_X/TSO_4 values is due to incorrectly specified $(NH_4)_2SO_4$ activities (`AWAS` variable in `BLOCK DATA ISO`) at RH = 33%. Both of these errors have been corrected in the version of ISORROPIA incorporated into CMAQ4.4. The `EX10` function call has been replaced with a slightly slower FORTRAN library routine and the $(NH_4)_2SO_4$ activities at RH = 33% have been reduced from 100 to 30. Using the revised ISORROPIA code, aerosol NO_3 remains below 0.05 μg m^{-3} for all values of NH_X/TSO_4 (see dashed line in Figure 2a).

A second family of numerical instabilities was identified based on CMAQ results posted to the Models-3 Listserve (Tong, 2004). Tong reported that a small change to Connecticut NO_X emissions in CMAQ4.3 yields a rather large (~1 μg m^{-3}) change in aerosol NO_3 concentrations in the Ohio Valley and in California. These peculiar results are produced because in the CMAQ advection algorithm, tiny numerical changes in pollutant concentrations may propagate across the domain in both upwind and downwind directions. When these tiny changes encounter a grid cell with certain combinations of temperature, RH, TNO_3, NH_X, and TSO_4, aerosol NO_3 and NH_4 concentrations are impacted due to ISORROPIA instabilities. The second family of instabilities occurs at different NH_X/TSO_4 ratios in CMAQ4.3 and CMAQ4.4 (see Figure 2b). Although these instabilities do not produce as large an impact on absolute aerosol concentrations as the first family of instabilities (compare vertical scales in Figures 2a and 2b), they warrant further exploration due to their potential to confound the assessment of emission control strategies.

The second family of ISORROPIA instabilities is encountered at low RH, where the aerosol water content is low and the aqueous-phase ion concentrations are quite high. Activity coefficient calculations under these conditions require several iterations for convergence and, if stopped prematurely, the solutions may be numerically unstable. These calculations may not be necessary if a solid phase were allowed to form but, in CMAQ, it is assumed that all particles are metastable aqueous droplets irrespective of the ambient RH. Recent laboratory studies by Martin et al. (2003) provide data on the crystallization RH of atmospheric sulfate-nitrate-ammonium mixtures. Crystallization data may be incorporated into atmospheric models as demonstrated by Colberg et al. (2003). This approach will be explored in a future version of CMAQ.
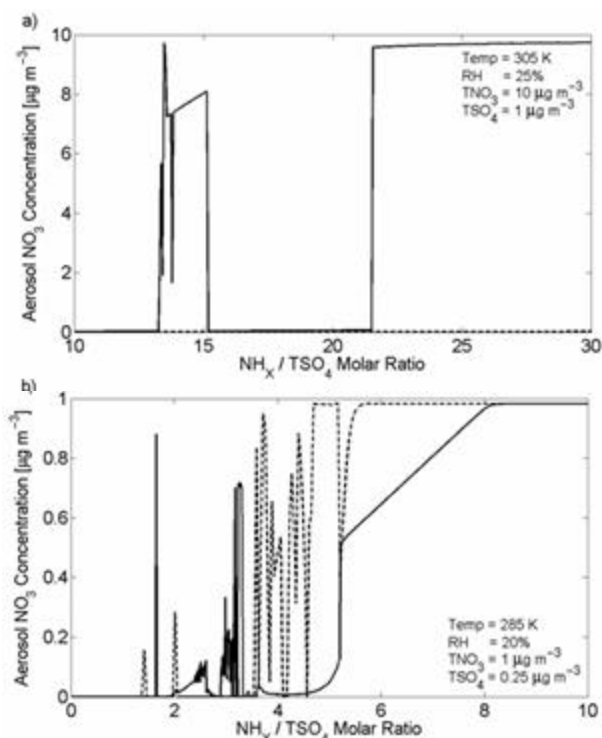

Figure 2. Examples of numerical instabilities in the ISORROPIA thermodynamics module released with CMAQ4.3 (solid line) and CMAQ4.4 (dashed line). Large errors that will occur in CMAQ due to infinitesimal changes in either NH_X or TSO_4 under the given meteorological conditions are illustrated by the sharp NO_3 gradients.

2.3 Code Structure and Documentation

Structural changes have been made to the CMAQ4.4 aerosol module in an effort to streamline the code. A new subroutine, `HCOND3`, has been added to calculate the second and third moment condensational growth rates. This replaces code that was formerly in subroutine `AEROSTEP`. In addition, subroutines `AEROPROC` and `AEROSTEP` have been merged into `AEROPROC` to remove a number of redundant variable declarations. The ENVIRON corporation reported an inconsistency in the organic aerosol molecular weights in subroutines `ORGAER3` and `RDEMIS_AE`. This is remedied in CMAQ4.4 and has minimal impact on the model results. A significant effort has been made to strengthen the in-line documentation in the aerosol code. Every major scientific formula in the aerosol module has been annotated with comments that permit users to trace the formula back to an equation in a journal article or published report where it is described thoroughly. These in-line documentation enhancements should aid the CMAQ users seeking to modify scientific algorithms within the model.

3. UPCOMING DEVELOPMENTS

Several aerosol module developments are anticipated in the 2005 public release of CMAQ. Heterogeneous interactions between gas-phase pollutants and sea-salt particles will be treated. A dynamic gas-particle partitioning algorithm is being explored currently, and may be included in the 2005 release if it is found to alter substantially the fine particle mass or composition relative to the instantaneous equilibrium assumption employed currently in CMAQ. The numerical instabilities displayed in Figure 2b will be removed from ISORROPIA, if possible. In addition, a second thermodynamic algorithm may be released with CMAQ to contrast the ISORROPIA results. Lastly, an aerosol mass conservation error identified by scientists at the Georgia Institute of Technology (Russell, 2004) is under investigation currently and should be resolved in the 2005 release.

Tools will be released in 2005 to enable users to probe the sources of modeled aerosol concentrations. With one tool, the contributions of different formation pathways to ambient sulfate concentrations can be determined. With a second diagnostic tool, users may calculate the contributions from individual source categories and/or geographic regions to primary carbonaceous aerosol.

4. CONCLUSIONS

As reviewed in this paper, the aerosol module has been updated in CMAQ4.4. The revisions are intended to improve computational efficiency, numerical stability, and code documentation, without altering the model results substantially. The efficiency improvements will be beneficial for regulatory applications in which numerous model simulations are required to determine the most effective air quality management strategies. The numerical stability improvements will assist with the interpretation of model results, through the mitigation of anomalous outputs obtained from earlier versions of ISORROPIA. Finally, new scientific advancements in the aerosol module and new diagnostic tools are anticipated for inclusion in the 2005 public release of CMAQ.

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