NEW FEATURES OF THE 2003 RELEASE OF THE CMAQ MODEL

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1. Introduction

The Community Multiscale Air Quality Model (CMAQ) has been extensively revised in 2003. Changes include: updated science, corrected implementations, efficiency enhancements, and bug fixes. The biggest changes involve aerosol modeling, particularly nitrate aerosols and secondary organic aerosols (SOA). Nitrate modeling was updated so it is consistent with the most recent literature and the SOA implementation was corrected to allow for reversible semi-volatility. These changes resulted in substantially lower concentrations of both aerosol nitrates and SOA. Minor changes have also been made to aqueous processes and dry deposition. The details of changes to chemistry/transport modeling are presented in Section 2.

There have been major modifications to improve model efficiency. A new fast gas phase chemistry solver, known as the Euler Backward Iterative (EBI) scheme, has been developed for the CB4 mechanism. Also, some of the fastest reacting species have been dropped from the transport processors. The time step for operator splitting has been revised to allow different advective time steps by vertical layer. Details of these changes are described in Section 3.

Note that other components of the CMAQ system such as SMOKE and MCIP have also been revised recently. Revisions to MCIP are especially important because serious errors in the processing of the horizontal and vertical wind fields have been corrected. Thus, one needs to use version 2.2 of MCIP (see MCIP release notes for details) for CMAQ modeling.

2. Chemistry/Transport Modeling Changes

2.1 Secondary Organic Aerosols (SOA)

The formation of secondary organic aerosols (SOA) in the atmosphere occurs via the oxidation of organic compounds to form semi-volatile gases that can subsequently condense on particles. Because of their volatility, these compounds are also capable of evaporating from the particle-phase back to the gas-phase as temperatures increases. The SOA gas-particle partitioning model used in CMAQ is based on the Secondary Organic Aerosol Model (SORGAM) described by Schell et al. (2001).

Two major problems with the SOA module contained in the 2002 release of CMAQ were identified. First, the manner in which the gas-particle partitioning model was implemented allowed for condensation of the semi-volatile gases to the particle phase, but not for the reverse process of evaporation. Second, the amount of semi-volatile SOA precursors formed from the atmospheric oxidation of some of the organic compounds of anthropogenic origin could be over-estimated in some cases.

In the previous version of CMAQ, the gas-phase components of the semi-volatile SOA precursors were represented by ten model species, and the corresponding particle-phase components of the semi-volatiles were lumped into the two generic anthropogenic and biogenic SOA species. In the new reversible version of the SOA model, the ten gas-phase species have been replaced by ten species that represent the sum of the gas-phase and the particle-phase concentrations. The representation of the anthropogenic and biogenic components of the SOA remains unchanged. Dry deposition velocities for the new semi-volatile species are computed as a mass-based, weighted averages of gas-phase and particle-phase deposition velocities based on the gas-particle partitioning.

The production of SOA from alkanes, alkenes, and aromatics has been updated. The vapor pressures of the semi-volatile products of alkene reactions have been revised such that alkenes are no longer considered a significant source of SOA. A review of emissions data shows that many compounds lumped into higher alkanes (ALK5 in SAPRC99 and HC8 in RADM2) do not form SOA precursors. Therefore, in the new release, the molar yields of the semi-volatiles produced by reactions of ALK5 and HC8 are multiplied by the average mole fraction of SOA producers that are lumped into these species. Also, the production rate of semi-volatile gases from high-yield aromatics (ARO1 in SAPRC99 and TOL in RADM2) has been adjusted downward to account for the fact that benzene, a non-SOA producer, is lumped into these mechanism species. No adjustment factor is needed for CB4 since benzene is not lumped into either of the aromatic compounds in that mechanism. The changes to alkanes and alkenes do not affect CB4 either since SOA is not produced from these species in CB4.

The molecular weights of the two semi-volatile SOA precursors produced by the oxidation of monoterpenes have been changed from 184.0 to 177.0 to be consistent with the molecular weight of the corresponding particle-phase SOA. In addition, changes have been made to the rate constants used in the CB4 and RADM2 mechanisms for the reactions of monoterpenes.

2.2 Nitrate Modeling

Starting with the 2002 CMAQ release the heterogeneous conversion of N_2O_5 to HNO_3 is modeled as a pseudo first-order loss process (i.e., N_2O_5--> $2HNO_3$), where the reaction rate is a function of the aerosol surface area, the N_2O_5 gas-phase diffusion coefficient, particle radius, and reaction probability. The amount of HNO_3 retained in the aerosol phase is determined by equilibrium considerations. With the implementation of the heterogenous pathway, the rate constants for the corresponding gas-phase hydrolysis reactions in CB4 and RADM2 were lowered to the value used in SAPRC99. The SAPRC99 rate constant is almost an order of magnitude lower and reflected a more recent estimate of the upper limit on the homogeneous, gas-phase conversion rate.

For the 2003 CMAQ release three major changes have been made. First, a units conversion error that caused the amount of HNO_3 produced by the pseudo first-order reaction to be a factor of 1.7 too high was corrected. Second, the reaction probability used in calculating the pseudo first-order rate constant for converting N_2O_5 to HNO_3 was changed from a fixed value of 0.1 after Dentener and Crutzen (1993) to a function of aerosol composition based on experimental results suggesting that the heterogeneous N_2O_5 conversion rate is inhibited by the presence of nitrates in the aerosol phase (Mentel, 2000). Following Riemer et al. (2003), the reaction probability varies between 0.002 and 0.02 as a function of the mass ratio of aerosol sulfate to the sum of aerosol sulfate and nitrate. The third change is to remove the homogeneous, gas-phase conversion of N_2O_5 to HNO_3 because evidence exists to suggest that the conversion of N_2O_5 to HNO_3 proceeds in hydrated aerosols as opposed to the gas-phase (e.g., Jacob, 2000). The net effect of all these changes is to significantly lower CMAQ predictions of aerosol nitrates under most conditions.

2.3 Clouds: Chemistry, Scavenging and Wet Deposition

Several modifications were made to the CMAQ cloud module. The most significant change is the calculation of the effective Henry's law values using dissociation equilibrium constants from Seinfeld and Pandis (1998) for species that do not participate in cloud chemistry (e.g. HCHO, HNO_2, HO_2). Accounting for dissociation of these species will increase their dissolution into the cloud and rain water thus increasing scavenging and wet deposition. Other changes include updates to the table of coefficients for calculation of Henry's law using data from Sanders (1999) and the addition of MEK, MVK, and METHACROLEIN to the table. Within the aqueous chemistry module, the section of the code that calculates the incremental time-step for the cloud was changed to be more stable to prevent division by zero when concentrations are extremely low.

Changes were made to the washout of coarse aerosol number concentrations (NUMCOR). Previously, the NUMCOR washout was scaled against the SO_4 removal rate---but SO_4 is also produced within clouds. Linking NUMCOR in this way created a problem for the diameter of coarse-mode particles, since the mass of coarse particles was washed out depending on the rainfall rate. To solve this problem, NUMCOR is now washed out in the same manner as the coarse mass. This should preserve the aerosol diameter during the washout process.

2.4 Dry deposition

There are two dry deposition models available in MCIP, M3dry, which includes direct linkages to the Pleim-Xiu land surface model (PX LSM) in MM5, and RADMdry, which was originally developed for the RADM model. Version 2.2 of MCIP includes updates to both dry deposition schemes with an emphasis on winter simulations including the effects of snow cover.

There are three new dry deposition species: N_2O_5, NO_3, and generic aldehyde. In addition, there were several species for which dry deposition velocities were previously computed in MCIP but were not used in CMAQ for dry deposition: PAN, HONO, CO, and Methanol. The dry deposition surrogates for all chemical mechanisms have been updated to include these species.

2.5 Corrections for vertical diffusion of aerosols

Previously aerosol species were treated as mass density units in the vertical diffusion processing. This is incorrect because vertical diffusion was developed for gas-phase concentrations in molar mixing ratio. We converted the aerosols to molar mixing ratio (ppmV) units, including emissions sources and dry deposition sinks. The vertical profiles are now more realistic, typically with higher concentrations at lower levels.

2.6 Re-ordered processes for operator splitting

Previously vertical diffusion was done after advection, mass-adjustment and horizontal diffusion in the time-splitting configuration for CMAQ. This was then followed by the cloud, gas-phase chemistry and aerosol processes. It is now felt that a better characterization of the time-splitting would be effected by doing the vertical diffusion of concentrations before advection, primarily to get emissions injected into the proper layers before advection takes place (emissions are injected in the vertical diffusion processing).

Preliminary tests indicate improvement in the characterization of time-splitting. Because of these changes, the couple/decouple processes are now called only in sciproc, and the decoupling of the average concentration array, AGRID, is no longer required.

3. Modifications to Achieve Improved Efficiencies

3.1 New Gas Phase Chemistry Solver

Previous releases of the CMAQ model have included a non-generalized but fast chemistry solver - the Modified Euler Backward Iterative (MEBI) method. The MEBI solver has proven to be significantly faster than the other chemistry solvers in CMAQ and quite accurate when bench marked against the high accuracy Gear method. The only major shortcoming is that it cannot be completely generalized to handle any chemical mechanism, but rather must be individually customized for each mechanism. Nevertheless, it has become the solver of choice in most model applications because of its speed and accuracy.

The MEBI solver is based on the methods described by Hertel et al (1993), except that the analytical solutions derived for the $NO/NO_2/O_3/O_3P$ and $OH/HO_2/HNO_4/HONO$ groups of species were replaced by numerical solutions as formulated by Huang and Chang (2000). Use of the numerical solution enhances generalization, but increases computational costs. This release includes a new EBI solver in which the numerical methods used for the two groups were replaced by the original analytical formulations of Hertel et al (1993) as applied to the CMAQ version of CB4. Tests conducted with EBI show it to be about 2 times faster than the MEBI solver while differences in model predictions were found to be minimal (e.g., maximum differences on the order of 1 ppb for ozone).

Currently, the CMAQ EBI solver has certain limitations. First, it can only be used with the CB4 family of mechanisms. Extensions to other mechanisms are planned, but are not yet available. Second, the EBI solver cannot be used to conduct integrated reaction rate (IRR) analysis because of the fairly large time step used in EBI. Integrated process rate (IPR) analysis can be invoked however. Finally, the EBI solver includes error control parameters that affect both speed and accuracy. A default set of error control parameters has been incorporated in the solver to enhance model speed while maintaining accuracy.

3.2 Elimination of transport for fast-reacting CB4 species

In previous releases of CMAQ, all of the species included in the gas-phase chemistry mechanism have been subjected to transport (i.e., advection and diffusion in the horizontal and vertical). Some of these species are at very low concentrations and react very rapidly however. If the fast reacting species concentrations are governed primarily by the concentrations of slower reacting species, it might be possible to forego transport of the fast reacting species, thereby reducing computational costs. Tests results indicate little loss in accuracy when transport is eliminated for the following CB4 species: O, O^1D, OH, HO_2, C_2O_3, ROR, CRO, TO_2, XO_2, and XO_2N. In fact, the differences were sufficiently small to negate the need to invoke steady-state approximations, thereby maximizing computational savings. As a result, the CB4 mechanism files have been configured to eliminate the transport of these 10 species in the latest release. The run-time savings, when aerosols are included, should be on the order of about 5%. Thus, when combined with the new EBI solver, run time reductions on the order of 20% can be realized as compared to using the MEBI solver and transporting all species.

3.3 Changes in Time-stepping

Previously, the time-step for operator splitting was determined by the shortest Courant-condition limited advective time-step anywhere in the 3-d domain. Thus, the synchronization time-step tends to be dictated by high winds aloft, leading to unnecessarily short time-steps for advection in the lower layers where wind speeds are usually much less. Since the region of greatest modeling interest is the planetary boundary layer (PBL), the algorithm was modified to try to get reasonable sync steps near the surface with equal advection steps and allow multiple advection steps aloft, if needed.

There is a new control parameter which specifies the model layers which determine the synchronization time-step. The new input parameter (SIGMA_SYNC_TOP) is the sigma value of the highest model layer to be used in determining the sync time-step. All model layers from the ground up to this sigma level will have the same advective time-step which is the minimum Courant-condition limitation in these layers. The model synchronization time-step is also set to this value such that these model layers will execute horizontal advection once per sync time-step. The default value for this parameter is 0.7 which reflects a height above ground of about 2.5 km. This is intended to include most of the daytime PBL in most areas. For model simulations where the maximum PBL height is expected to be very different from this height (e.g. dry climates and/or high elevations) the user can specify a different value for this parameter. The ZF parameter on the METCRO3D file from MCIP can provide guidance on the heights of the sigma layers.

There also has been a modification of model defaults for maximum and minimum synchronization time-step values. On the basis of model testing with 32 km grid resolution compared to benchmark runs with 6 minute synchronization time-steps, the maximum recommended (default) sync time-step is now set to 12 minutes rather than the previous 15 minutes. Also, based on testing at 2 km grid resolution, the recommended (default) minimum sync time-step is now 1 minute rather than the previous 5 minutes.

Initial tests compared with fixed, short synchronization steps show an improvement in the accuracy in the lower layers. Also, there is some decrease in CPU time for coarser resolutions, but the opposite is sometimes true for the finer resolutions.

3.4 Parallel Processing Changes

When the code runs in parallel in a so-called MPICH cluster, synchronization between the node that launches the run and the other participating nodes can be a problem, evidently due to network file system (NFS) latency. A spin-wait code was employed to attempt to deal with this problem by delaying execution for 60 sec. after processor 0 issued an IO/API call to open a new CONC file. However, the spin-wait solution allows incorrect IO/API header data to be written to the output CONC and ACONC files. Specifically, the netCDF TFLAG variable, representing the timestamp (record), can have invalid or no data at the first date/time for a number of file variables. A correction obtained from Zion Wang of CERT-UC-Riverside has been applied, which involves simplifying the opening and closing of the output concentration file for the case that the file does not previously exist. The problematic spin-wait code is no longer used.

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