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COUPLED METEOROLOGY AND CHEMISTRY WITHIN GEM-MACH: INVESTIGATING FEEDBACKS WITH CANADA'S AIR QUALITY FORECAST MODEL

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1. INTRODUCTION

Environment Canada is a participant in 2nd phase of the Air-Quality Model Evaluation International Initiative (AQMEII). In this phase of the multi-model comparison, coupled models are the focus, and specifically, fully coupled models, in which the chemical and meteorological components are allowed to interact. Environment Canada's operational air-quality model GEM-MACH is on-line, but only partially coupled, in that the chemistry portions of the model are influenced by meteorology, but feedbacks from chemistry to meteorology are presently not allowed. Progress towards incorporating feedbacks between chemistry and weather in GEM-MACH will be described in this talk.

2. GEM

2.1 GEM Overview

The Global Environmental Multiscale (GEM) model is the source of the Canadian government's operational weather forecast. GEM makes use of a rotated latitude–longitude coordinate system for its horizontal domain, with global uniform (GU), global variable (GV), and local area model (LAM) configurations. The grid resolutions used in GEM vary from 2 degrees (GU) to 0.0225 degrees (~ 2.5km, LAM). GEM was first described in Côté *et al.*, (1998), though many modifications to the model code have taken place since that time. The version of the model to be examined here makes use of a subset of GEM's regional North American forecast grid, with a resolution of 0.1375 degrees (~15.3 degrees), operating on a time step of 450s (7.5 minutes). In the work carried out here, GEM version 3.3.6 with physics version 4.7.2.1 (GEM-MACH version 1.4.6) of the model was used, with modifications to include 58 hybrid-coordinate levels from the Earth's surface to 10 hPa, with layer thickness increasing monotonically with height (the first three levels above the surface, relative to standard pressure at sea level are at 42, 126, and 225 m), and the incorporation of the most recent version of the Milbrandt-Yau bulk microphysics scheme (Milbrandt and Yau, 2005). In the operational forecast cycle, objective analyses at 0, 6, 12 and 18 UT are used to drive the GU configuration of the model, the results of which along with regional objective analyses are used to drive the North American LAM forecast. Hourly piloting files from the latter are used to drive local 2.5km resolution LAM simulations.

2.2 Existing GEM parameterizations making use of chemical variables.

Chemical variables are specified essentially as numerical constants within the standard GEM, in two sections of the model; the cloud microphysics scheme, and the radiative transfer processes. These will be described in more detail here, given their relevance to the conversion of the model to a fully coupled status.

The Milbrandt-Yau 2-moment bulk microphysics scheme solves prognostic equations for the total number concentration and the mass mixing ratio of six hydrometeors (cloud droplets, rain, ice, snow, graupel and hail). The size distribution of the hydrometeors is parameterized using integrated distribution functions (moments) where the size distribution is given by

$$N_x(D) = N_{0x} D^{\alpha_x} e^{-\lambda_x D} \qquad (1)$$

With the p'th moment described by:

$$M_x(p) \equiv \int_0^\infty D^p N_x(D)dD = N_{0x}\frac{\Gamma(1+\alpha_x+p)}{\lambda_x^{p+1+\alpha_x}} \qquad (2)$$

The total number concentration of each of the six hydrometeors is the zero'th moment:

$$N_{Tx} \equiv \int_0^\infty N_x(D)dD = M_x(0) \qquad (3)$$

The mass mixing ratio of each of the six hydrometeors is the third moment:

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$$q_x \equiv \frac{\pi\rho_x}{6\rho}\int_0^\infty D^3 N_x(D)dD = \frac{\pi\rho_x}{6\rho}M_x(3) \qquad (4)$$

The scheme solves the system of 12 differential equations ($\frac{dq_x}{dt}, \frac{dN_{Tx}}{dt}$ x=1..6), including terms for eight microphysical processes (collection(accretion), conversion, freezing, nucleation, ice multiplication, melting, vapour diffusion and sedimentation). The impact of aerosols on the cloud microphysics scheme is through the nucleation of cloud droplets. Supersaturation is diagnosed as a function of the updraft speed, temperature and pressure, and the number of cloud droplets at time t is given by

$$N_c^t = \max\left[N_c^{t-1}, N_{CCN}(ssat)\right] \qquad (5)$$

The number of cloud condensation nuclei at supersaturation is taken from two simple functions, one for continental, the other for maritime airmasses, following Cohard et al. (1998), Figure 1.

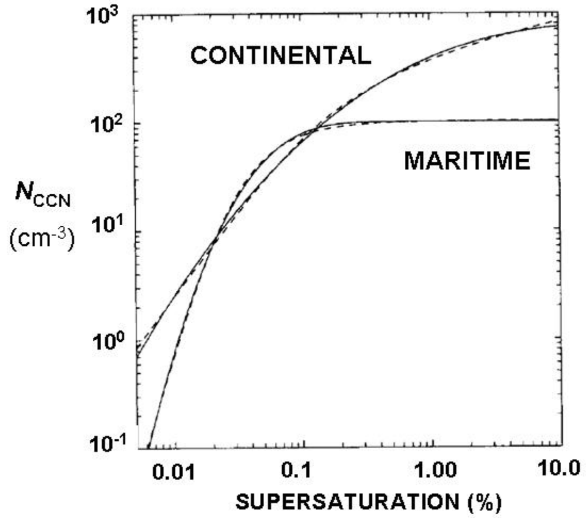

Fig. 1. Empirical relationship between supersaturation and number of cloud condensation nuclei in GEM, after Cohard et al (1998).

One potential point of connection to between the meteorology and the chemistry is thus via the characterization of the CCN (aerosols) and cloud droplet nucleation.

The radiative transfer algorithms of GEM make use of both aerosol and O_3 in two locations, with specified concentration profiles. The algorithms make use of the following function of the background normalized incremental aerosol optical depth for solar and longwave at each gridpoint within a given x,z slice:

$$\tau_{ae}(x,z) = \kappa\left[\left(\delta - \frac{2\varepsilon}{\pi}\left|\ell_x\right|\right)\frac{\Delta z_{x,z}}{\sum_z \Delta z_{x,z}}\right] \qquad (6)$$

Where $(\kappa, \delta, \varepsilon)$ are (0.730719, 0.25, 0.2) for continental airmasses, and (0.912819, 0.13, 0.1) for oceanic airmasses (Toon and Pollack, 1976), and ℓ is the latitude. The values of the aerosol optical properties of single scattering albedo, and asymmetry factor are set to (0.872212, 0.647596) for continental air and (0.982545, 0.739002) for maritime air, for four shortwave bands (longwave effects are small were neglected). The aerosol in the above parameterization is assumed to be uniformly distributed over a layer extending from the surface to a height of 1500m above the surface. The local aerosol optical depth and the assumed optical properties of the aerosol, are thus the second point at which chemical variables may affect the meteorology.

Ozone is specified in the operational GEM using a latitude and height 2D monthly climatology from Fortuin and Kelder (1998), extending from the surface to 0.3 mb. The ozone from within the heretofore decoupled chemistry may thus be used in the place of the climatology, allowing a further feedback to the chemistry.

3. GEM-MACH

3.1 GEM-MACH overview {from AURAMS}

GEM-MACH refers to the combined model comprising GEM as described above, and the Modelling Air-quality and Chemistry module, a subroutine package called from within the GEM physics module. The on-line framework under which GEM-MACH was constructed allows for full coupling of the meteorology, though the present operational version only allows one-way information transfer, from the meteorology to the chemistry. The transport of tracers is handled within GEM prior to the chemistry (3D semi-Lagrangian advection, with a cubic Lagrange interpolant). The remaining operators for the chemical variables are handled within the chemistry algorithms with one-step forward operator splitting. The chemical operators include:

(1) Major point source plume rise (plume rise buoyancy calculations according to Briggs, 1984, Briggs, 1985, Turner, 1985 and Sharf *et al*, 1993),

(2) Vertical diffusion (fully implicit Laasonen approach (Richtmyer, 1994), with area source emissions and gaseous deposition incorporated

either as boundary conditions on the diffusion equation or as a separate operator.

(3) Gas-phase chemistry calculations follow (42 gas species, ADOM-II mechanism; Stockwell and Lurmann, 1989, numerically solved using the method of Young and Boris, 1977), with secondary organic aerosol condensable mass being estimated using the approach of Odum *et al* (1996), with updated organic aerosol yields.

(4) The Canadian Aerosol Module (Gong et al, 2003), which includes particle microphysics (condensation of sulphate and secondary organic condensable mass, coagulation, nucleation of sulphate aerosols), aqueous-phase chemistry including aerosol activation (Gong et al, 2006) (ADOM aqueous phase mechanism, Venkatram *et al*., 1988, Fung *et al*., 1991; using the solver of Young and Boris, 1977, for integration), inorganic heterogeneous chemistry of sulphate, nitrate and ammonium (HETV solver, Makar et al, 2003), sea-salt emissions (Gong *et al*., 2003), and finally particle settling and deposition (Gong *et al*., 2003, Gong *et al* 2006.).

Particles are chemically resolved in GEM-MACH using 9 species (sulphate, nitrate, ammonium, secondary organic, primary organic, black carbon, crustal material, sea-salt, and water). The model uses a sectional approach to resolve the particle size distribution (choice of 2 or 12 bins). Some of the chemistry processes subdivide the bins into sub-bins in order to better capture size-dependant microphysical processes.

3.2 GEM-MACH parameterizations for feedback-relevant processes

The chemical algorithms make use of meteorological inputs such as temperature, wind speed, relative humidity, etc., to drive the processes described above. Some processes within the operatonal GEM-MACH are simplifications which could also be more directly connected to the meteorology.

The photolysis rates in the model are derived from height versus solar zenith angle tabulated values of $O_3 + h\nu \rightarrow O(^1D)$ and $NO_2 + h\nu$ rates, with solar zenith angle dependant scaling factors for the other species. A cloud photolysis correction is carried out based on the location within, above, or below clouds. One possible improvement would be to derive photolysis rates directly from the 12 wavelength band (9 long-wave, 3 short-wave) radiation code of the meteorology model. .

Aerosol activation (i.e. droplet nucleation) is treated within GEM-MACH's chemistry module in order to determine the part of the aerosol spectrum that will participate in cloud processing (uptake of gases, aqueous-phase reactions, removal through precipitation production and below-cloud scavenging, precipitation evaporation, etc.; Gong *et al,* 2006). Meteorological variables such as the cloud liquid water content, cloud fraction, cloud to rain conversion rate (autoconversion/precipitation production), and precipitation evaporation from the meteorological code are all made use of in GEM-MACH's cloud processing.

The current aerosol activation scheme within GEM-MACH's chemistry module differs from the current cloud droplet nucleation parameterization in GEM's physics module (eqn 5), with the use of the Jones et al (1994) empirical relationship:

$$N_d = 375(1 - \exp(-2.5 \times 10^{-3} N_a)) \qquad (7)$$

where N_d and N_a are the droplet number and aerosol number concentrations respectively. The particles which activate are determined by counting downwards from the largest size bin in the aerosol distribution towards the smallest size bin until the total N_d (from eqn 7) is reached.

Recent off-line tests within the AURAMS model (Gong et al, 2012), have suggested that the more accurate spatial distributions of cloud water may be achieved through the adoption of the the more physically-based activation scheme of Abdul-Razzak and Ghan (2002). In this methodology, aerosol activation is determined by comparing the upper and lower bounds of critical supersaturation of each size bin (S_i) to the maximum supersaturation in an updraft (S_{max}), through a number (N_i)-weighted effective critical supersaturation (S_e):

$$S_{\max} = \frac{S_e}{\left[0.5\left(\frac{\varsigma}{\eta}\right)^{3/2} + \left(\frac{S_e^2}{\eta + 3\varsigma}\right)^{3/4}\right]^{1/2}} \qquad (8)$$

and

$$S_e^{2/3} = \frac{\sum_{i=1}^{nb} N_i}{\sum_{i=1}^{nb}\left(N_i / S_i^{2/3}\right)} \qquad (9)$$

where ς and η are parameters dependant on updraft velocity, growth coefficient (accounting for diffusion of heat and moisture to particles), surface tension, etc., and S_i depends on size, hygroscopicity, and surface tension characteristics of the particles in a given section/bin.

4. CURRENT WORK

The conversion of the on-line but decoupled GEM-MACH to a coupled model is in progress, following the stages outlined below.

(1) The current cloud droplet nucleation scheme in GEM's physics module (eqn 5) will be replaced with the current activation scheme in the GEM-MACH chemistry (eqn 7), using the on-line aerosol number concentration as a first step to explore the on-line aerosol feedback to cloud microphysics and the resulting cloud fields. The cloud droplet number concentration so determined in GEM's physics will be passed to GEM-MACH's chemistry for cloud processing calculations.

(2) The more physically-based Abdul-Razzak and Ghan scheme (eqn 8, 9) will be implemented directly in the GEM physics module, making use of the on-line size and chemically resolved aerosol information and again the resulting cloud droplet number concentration will be used in GEM-MACH's cloud processing calculations.

(3) GEM's existing continental versus marine, constant AOD parameterization (6) will be replaced by local size and speciation dependant calculations making use of the GEM-MACH particle composition and size distribution. Both empirical (IMPROVE, Malm et al, 1994) and Mie calculation options may be used for this purpose.

The IMPROVE formula for aerosol scattering and absorption may be used to calculate the net extinction coefficient at each model layer:

$$b_{sc,ab}(x,z) = \left\{ \begin{matrix} 3 f_T(RH)\left[\left(AMSUL\right)+\left(AMNIT\right)\right] \\ +4\left[OM\right]+\left[Soil\right] \\ +0.6\left[Course\right]+10\left[EC\right] \end{matrix} \right\}_{x,z} \tag{10}$$

Where RH is the relative humidity, AMSUL and AMNIT are the model-derived concentrations of ammonium sulphate and nitrate, and OM, Soil, Course and EC are the concentrations of organic matter, soil, course particulate matter and elemental carbon, respectively. The values of $\tau_{ae}(x,z)$ in (6) may thus replaced by

$$\tau_{ae}(x,z) = b_{sc,ab}(x,z) * \Delta z_z \tag{11}$$

As noted under (6), a fixed value of the single scattering albedo (fraction of total extinction due to scattering) is employed within the operational GEM. From equation (10), noting that elemental (ie. light absorbing) carbon is the only absorbing component, the value of the single scattering albedo is given by:

$$\Omega = 1 - \frac{10\left[EC\right]}{b_{sc,ab}(x,z)} \tag{12}$$

A more numerically complex methodology would be to make use of a Mie scattering algorithm. One complication here is that a finely resolved size distribution is required to carry out Mie calculations

Each of these stages is being tested with short runs of the meteorological model, and progress on the tests will be presented at the Conference.

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