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# OPERATIONAL AIR QUALITY FORECASTING IN CANADA: NUMERICAL MODEL GUIDANCE FOR GROUND-LEVEL OZONE AND PARTICULATE MATTER

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## 1. INTRODUCTION

It is becoming more and more evident that poor air quality, as indicated by high ambient levels of ground-level ozone and particulate matter, has a significant impact on human health (e.g., Brook et al, 2002). On a day-to-day basis, there are segments of the population that may be impacted significantly even by moderately poor air quality.

In order to assist the Canadian populace in making individual decisions to alleviate the effects of poor air quality, the Meteorological Service of Canada (MSC) initiated in the summer of 1999 an Air Quality Forecasting program for eastern Canada. This program, initially limited to a forecast of ground-level ozone, was carried out in collaboration with the eastern Canadian provinces and other interested stakeholders. The main numerical guidance for such a forecast was provided using a source-oriented photochemical oxidant model, the Canadian Hemispheric and Regional Ozone NOx System (CHRONOS) as the chemical transport model (CTM) (Pudykiewicz, et al., 1997; Sirois et al., 1999). Following the pilot study in eastern Canada, the Air Quality Prediction Program was extended to cover all of subarctic Canada in 2001.

## 2. THE METEOROLOGICAL MODEL

The meteorological model that supplies CHRONOS with the necessary meteorology is the Canadian operational regional version of the Global Environmental Multiscale (GEM) model. It is a global 28 eta level model with a constant 24 km grid covering North America and adjacent waters. (For details, see http://www.msc-smc.ec.gc.ca/cmc/op_systems/regional_forecast_e.html ) It is integrated to 48 hrs with a time step of 720 seconds with hourly outputs for CHRONOS.

## 3. EMISSIONS INVENTORY

In addition to meteorological input files, CTMs such as CHRONOS require detailed emission input files that describe the time-varying emissions of each chemical species considered by the CTM at each model grid cell (e.g., Russell and Dennis, 2000). The primary data sets used to create the emission input files needed by CHRONOS were the 1990 Canadian and U.S. "criteria air contaminant" (CAC) emission inventories. Annual emissions for North America for six pollutant "species" ($SO_2$, $NO_x$, VOC, $PM_{2.5}$, $PM_{10}$, and CO) are available by province in the 1990 Canadian CAC emissions inventory (Deslauriers, 1996) and by state and county in the 1990 U.S. inventory (U.S. EPA, 1993). The U.S. 1990 national CAC inventory also contains $NH_3$ emissions.

These two data sets, however, only contain emissions reported on an annual basis for a small set of aggregated species (e.g., $NO_x$, VOC) for political jurisdictions rather than for grid cells. In order to bridge these differences, a software tool called an emissions processing system must be used (e.g., Dickson and Oliver, 1991; Houyoux et al., 2000). The three main processing steps that must be performed by this tool are temporal disaggregation, chemical speciation, and spatial disaggregation. Chemical speciation is particularly important for volatile organic compounds (VOC), where the inventories report only total VOC emissions but CTMs require more detailed emissions information for individual VOCs or groups of VOC species.

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The Canadian Emissions Processing System (CEPS: see Moran et al., 1997; Scholtz et al., 1999) was used to process the two national 1990 inventories on a season-day basis, where the seasons have been defined to be three calendar months each, with winter spanning the December-February period, spring the March-May period, etc. Speciated hourly emissions were estimated for 9 inorganic species (NO, $NO_2$, $SO_2$, $SO_4$, CO, $NH_3$, $PM_{2.5}$, $PM_{10}$, and HONO) and 11 single or "lumped" organic species associated with the ADOM-II gas-phase chemical mechanism (e.g., Pudykiewicz et al., 1997). The domain that has been used by CEPS for processing the 1990 Canadian and U.S. inventories for this assessment is illustrated in Figure 1. The map projection used is a polar stereographic projection, true at 60°N, with the vertical axis aligned with the 100°W meridian. The grid spacing is 21 km.

CEPS also separated the anthropogenic emissions in the two national inventories into four primary source types: major point sources; minor point sources; area and off-road mobile sources; and on-road mobile sources. This partitioning gives greater flexibility for making any adjustments that are required to the emission files. Biogenic emissions, on the other hand, which vary strongly with meteorological conditions and season, were not treated by CEPS, but rather were estimated "on the fly" in CHRONOS using the BEIS2 algorithm (Pierce et al., 1998). Surface-temperature and photosynthetically-active-radiation values required by BEIS2 are obtained from GEM fields.

## 4. THE 2002 AIR QUALITY PREDICTION PROGRAM

### 4.1 The Chemical Transport Model

The national air quality forecast at the Meteorological Service of Canada (MSC) was launched in May of 2001. The essential component of the air quality prediction system is CHRONOS which is a multiscale air quality model initially designed for the prediction of tropospheric ozone (Pudykiewicz et al., 1997). The model simulates dispersion and complex chemistry of both nitrogen oxides and volatile organic species using inventories of actual anthropogenic emission. The model is also equipped with an online package to calculate biogenic emissions based on vegetation data. Some points of note are:

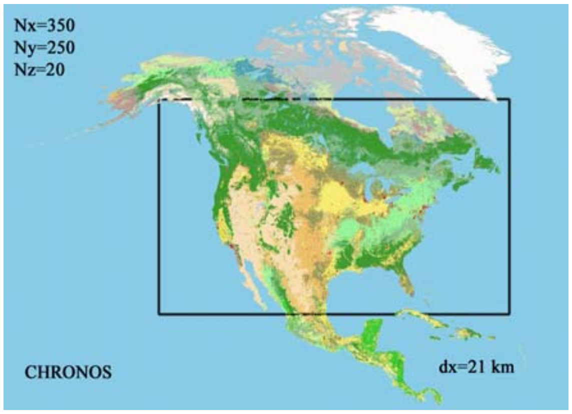

Figure 1: Continental domain of the operational CHRONOS. Horizontal grid spacing is 21 km.

- The forecast of chemical fields is performed on a domain containing most of the pollution sources in Canada and the USA
- The resolution of the forecast of chemical variables is sufficient to forecast regional scale air quality
- CHRONOS also provides a good air quality forecast on the scale of a large city agglomeration
- Computing aspects: large number of degrees of freedom, time of execution of the order of 50 minutes per day of simulation (on one processor of a NEC SX–5 supercomputer)
- CHRONOS is used extensively to investigate smog episodes in North America.

The current 2002 version of CHRONOS includes the parameterization of unresolved mixing, wet scavenging of tracers and a bulk aerosol scheme.

### 4.2 Experimental Bulk Aerosol Scheme

In recent years, a significant scientific effort has been dedicated to the modelling of aerosols both in climate and in air pollution research. From the mathematical point of view, the problem is governed by the continuity equation with terms representing coagulation, condensation and emissions, which describes the evolution of the aerosol number concentration in a four-dimensional space.

For the summer 2002 prediction program, an experimental particulate matter (PM) component was added to the model. The PM component consisted of bulk $PM_{2.5}$ and $PM_{10}$ made up of four chemical components: primary PM due to direct emissions (Section 3); secondary sulphate produced by gas-phase reactions; secondary organic aerosol based on a

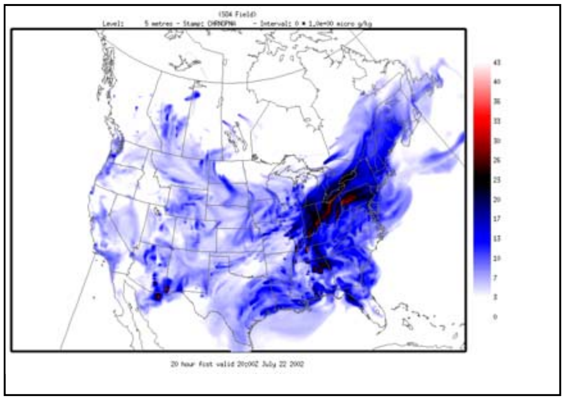

Figure 2: Bulk concentration of sulphate aerosol predicted by CHRONOS.

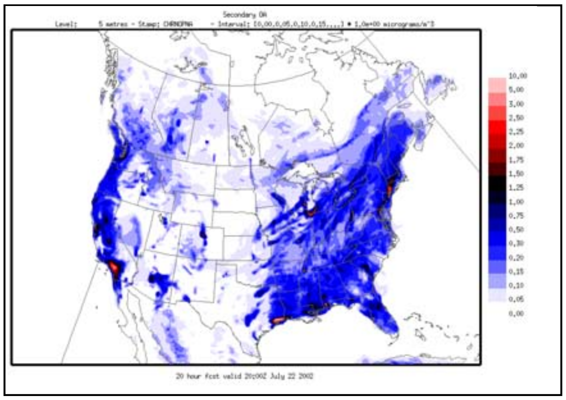

Figure 3: Bulk concentration of Secondary Organic Aerosol (SOA) predicted by CHRONOS.

parameterization of Pandis et al. (1992); and condensed water (see Section 4.2). Figures 2 and 3 show sample fields for two of these chemical components. Note that this simple treatment does not represent all of the sources of PM mass (e.g., Section 4.4), but it includes both primary and secondary sources and does not add significantly to the computational burden. In the future, the method of moments may be used to represent the PM size distribution.

### 4.3 Aerosol Water in CHRONOS

The liquid water contained in atmospheric particles is an important constituent of the total particle mass. At high relative humidities, the mass of aerosol-bound water can, for example exceed the mass of inorganic constituents. A methodology for calculating the mass of water as one of the components of the forecast aerosol is therefore required.

The amount of water contained in atmospheric particles is a complex function of the relative proportions of the other particle constituents, both inorganic and organic. The content of an inorganic aerosol may be determined via the iterative solution of the system of equations describing the equilibrium reactions between the inorganic gases (primarily ammonia and nitric acid in polluted environments) and the ions and crystalline solids within the particles (cf. Nenes et al., 1998). The water content of liquid-phase organic aerosols may be determined through the use of an iterative version of Raoult's Law as described by Pankow (1994a,b), making use of group theory to describe the activity coefficients of the organic mixture (Sandler, 1999). While both approaches will yield estimates of particle-bound water content (and may be combined iteratively to approximate the water content of a heterogeneously mixed aerosol), they pose severe processing time and memory constraints on an operational PM prediction. Explicit composition information is required for these methods, with each aerosol chemical component to be resolved requiring additional memory storage space. The explicit composition calculations are iterative. Estimates of total forecast processing time using models with detailed heterogeneous chemistry (e.g. Moran et al., 1998) show that these iterative calculations may consume the bulk of the processing time required for a given simulation. The processing demands may be sufficiently high to prevent the creation of a new forecast within an operational timeframe.

Non-explicit-composition, empirical methods for estimating aerosol water are also available, and due to the operational constraints for the current version of CHRONOS, a method of this nature has been used (Hänel, 1976; Gong et al, 2002). Relative humidity is related to the aerosol size via a form of the Köhler equation:

$$\mathrm{RH} = \exp\left( -\overline{\Phi}\,\overline{\nu}\,\frac{M_w}{\overline{M}_s}\frac{m_s}{m_w} \right) \exp\left( \frac{2\sigma}{\rho_w R_w T r_d}\frac{1}{f_r} \right)$$

where RH is the relative humidity (0 to 1 scale), $\overline{\Phi}$ is the empirically derived osmotic coefficient for the mixture, $m_s$ and $m_w$ are the masses of solute and water, $M_w$ and $\overline{M}_s$ are the molecular mass of water and the average molecular mass of the solute mixture, $\sigma$ is the surface tension between the solution droplet and air, $R_w$ is the gas constant for water vapour, $r_d$ is the dry aerosol radius, T is the temperature, and $f_r$ is the radius ratio (wet to dry; $f_r = r/r_d$, where r is the wet aerosol radius). Details of the implementation of the above equation may be found in Gong et al. (2002).

In the current implementation of CHRONOS, particulate matter is carried in bulk mode; the water calculations are performed using an assumed log-normal distribution, as is done in the particle deposition. Bulk particle sulphate and organics are distributed log-normally on input to the water calculation routine over 12 size bins. These values are used in conjunction with the above equation to determine the water content in each of the assumed bins. The resulting water amounts are aggregated into "Fine" and "Coarse" modes, based on the initial distribution.

### 4.4 Cloud and Aqueous-Phase Chemistry

Clouds play an important role in the processing and cycling of chemicals in the atmosphere. They affect the distribution and composition of atmospheric chemicals through modification of photolysis rates, redistribution of tracers by vertical transport/mixing, removal by wet deposition, and production/transformation by aqueous-phase reactions. For example, it is believed that the most significant contribution to the production of atmospheric sulfate aerosols is due to aqueous phase oxidation in cloud droplets. The current CHRONOS has a simple representation of cloud shading (impact on photolysis rates). A parameterization of wet removal of species is also included. However aqueous-phase chemistry (mass transfer and oxidation) is not yet represented in the current CHRONOS, which could result in a significant under-prediction of atmospheric sulfate aerosols. The inclusion of aqueous-phase chemistry in the air quality forecast model could entail a significant increase in computational burden due to the stiff nature of the system. In the context of a new Unified Regional Air Quality Model being developed at the Meteorological Service of Canada (Moran, et al., 1998), the cloud process and aqueous-phase chemistry component are currently being evaluated (see Gong et al., 2003). Significant code optimization will be required before it can be implemented in the operational forecast model.

## 5. PERFORMANCE EVALUATION

The regional-scale version of CHRONOS was extensively evaluated in the past under the auspices of North American Research Strategy for Tropospheric Ozone NARSTO. Model evaluation for the initial version of CHRONOS, including an analysis of scattergrams, time series, spectral analysis, is discussed in detail in Sirois, et al. (1999). The current high-resolution operational model is the subject of a similar, intensive evaluation. Figures 4-7 and Table 1 present some examples of the 2002 model simulations and model performance with respect to tropospheric ozone.

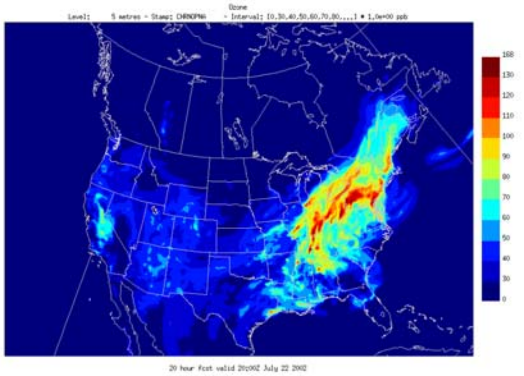

Figure 4: Ozone episode simulated by CHRONOS on 22 July 2002, 1500 EDT.

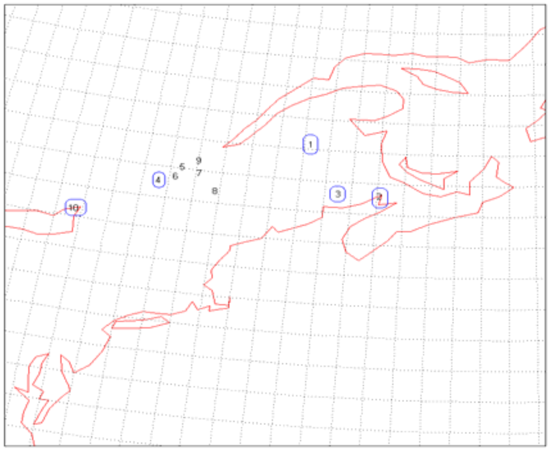

Figure 5: Location of stations used for the demonstration of the model results.

Table 1: Correlation of observed and predicted hourly surface ozone concentrations for five stations in eastern Canada for the July 9 – September 3, 2002 period.

| Station | Correlation coefficient |
|---|---|
| 1 | 0.73 |
| 2 | 0.80 |
| 3 | 0.74 |
| 4 | 0.80 |
| 10 | 0.85 |

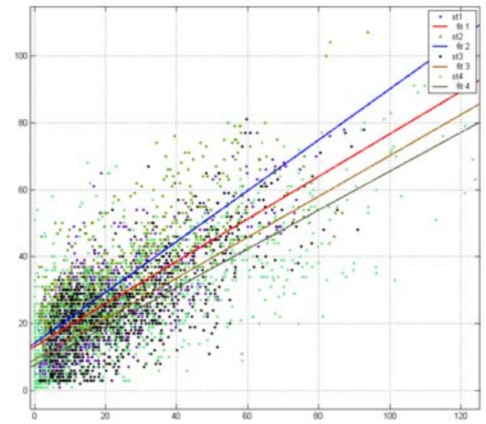

Figure 6: Scattergrams for stations 1-4 on Figure 5 and linear fits; model results are on the horizontal axis and observations on the vertical axis (station 4 is located in Montreal).

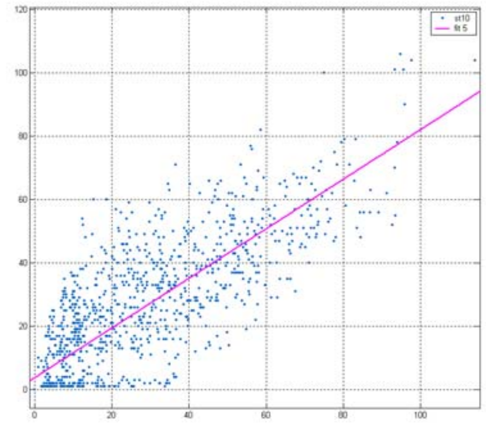

Figure 7: Same as Figure 6 but for station 10 (Kingston, On)

## 6. CONCLUDING REMARKS

The 2002 prediction program used gridded emissions based on the 1990 emissions inventory. Work is currently underway to update this emission inventory. Tropospheric ozone levels obtained as output from CHRONOS are used as important guidance in the issuance of air quality forecasts in the various regions and provinces of Canada (Maps of predicted ozone concentration generated by the CHRONOS system can be viewed at http://www.msc-smc.ec.gc.ca/aq_smog/chronos_e.cfm). Since these model outputs are used in real-time by meteorologists in the regional forecast offices across Canada, immediate feedback can be provided on the performance of the model, especially during smog episodes. Furthermore, a systematic model performance evaluation is done at the end of each summer by comparing the model predictions with observations collected by ground-level ozone measurement networks.

For the summer 2002 prediction program an experimental particulate matter (PM) component was added to the model. The PM component consisted of bulk PM2.5 and PM10, made up of primary components, secondary sulphate, secondary organic aerosol, and condensed water.

While continent-wide maps of the tropospheric ozone component are available on the web for public access, the PM components are being evaluated internally by Environment Canada during the summer 2002 before making them publicly available.

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