2.3 CLOUD PROCESSING OF GASES AND AEROSOLS IN A REGIONAL AIR QUALITY MODEL (AURAMS) AND ITS EVALUATION AGAINST PRECIPITATION-CHEMISTRY DATA

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1. INTRODUCTION

Clouds play an active role in the processing and cycling of chemicals in the atmosphere. Gases and aerosols can enter cloud droplets through absorption/condensation (of soluble gases) and activation and impact scavenging (of aerosol particles). Once inside the cloud droplets these tracers can dissolve, dissociate, and undergo chemical reactions. For example, it is believed that aqueous phase chemistry in cloud is the largest contributor to sulphate aerosol production. Some of the aqueous-phase tracers will be removed from the atmosphere when precipitation forms and reaches the ground. However, the majority of clouds are nonprecipitating, and upon cloud dissipation and evaporation, the tracers, physically and chemically altered, will be released back to the atmosphere. Updrafts and downdrafts in convective clouds are also efficient ways of redistributing atmospheric tracers in the vertical. It is therefore important to represent these cloud-related physical and chemical processes when modelling the transport and transformation of atmospheric chemical tracers, particularly aerosols.

A new multiple-pollutant (unified) regional airquality modelling system, AURAMS, with size- and chemical-composition-resolved aerosols is being developed at the Meteorological Service of Canada (Moran et al., 1998). In the current version of AURAMS, many of the cloud processes mentioned above are represented. Initial evaluations of AURAMS are being carried out for two summertime multi-day simulations in August 1988 and August 1989 over eastern North America (during the Eulerian Model Evaluation Field Study [EMEFS] period; Hansen et al., 1991). This study will focus on the impact of the various cloud processes on modelled regional aerosols and an evaluation against the available precipitation-chemistry data for the first period. In what follows we will first give a brief description of the various cloud processes represented in AURAMS. We will then present the simulation for the period of August 1 to 6, 1988 – a hyper intensive observational period during the first EMEFS field campaign, illustrating the impact of cloud processes on modeled regional aerosols. Comparison of modelled tracer concentration in precipitation against observations will be discussed.

2. CLOUD PROCESSES AND THEIR REPRESENTATION IN AURAMS

The various cloud processes currently represented in AURAMS include activation (nucleation) scavenging of aerosols, aqueous-phase chemistry (both mass transfer and oxidation), and wet deposition. Descriptions for other components and processes represented in the current AURAMS can be found in Moran et al. (1998), Gong et al. (2002), Makar et al. (2002), and Zhang et al. (2002).

2.1 Aerosol activation (nucleation scavenging)

Aerosols (particularly those that are water soluble or contain water soluble components) can serve as cloud condensation nuclei (CCN) and, under suitable supersaturation conditions, condensation of water vapour on these CCN can proceed rapidly to lead to the formation of cloud droplets, i.e., aerosol activation process. The capture of aerosols in cloud droplets due to this process is often referred to as activation (or nucleation) scavenging. Activation can be determined from aerosol size, composition, and supersaturation. In the current version of AURAMS,

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the aerosol activation, inherited from the Canadian Aerosol Module, CAM (Gong et al., 2002), is based, however, on a simple empirical relationship between the aerosol number density $N_{aerosol}$ (cm⁻³) and the number density of cloud droplets $N_{droplet}$ (cm⁻³) formed on the activated aerosols (CCN) described in Jones et al. (1994):

$$N_{droplet} = 375(1 - \exp(-2.5 \times 10^{-3} N_{aerosol}))$$

Under this parameterization, the largest aerosol particles are assumed to activate first and the critical radius is determined by counting downward from the top end of the aerosol size spectrum until the total number of droplets is accumulated. The bulk cloud water content from the meteorological driver model is distributed evenly amongst the activated aerosols, and the cloud droplet size is then determined for each activated aerosol bin. This parameterization results in a relatively narrow cloud-droplet spectrum.

2.2 Cloud chemistry

Cloud/aqueous-phase chemistry usually refers to several processes including mass transfer between the gas and aqueous phases (cloud droplets). dissociation/ionisation of certain dissolved species, and chemical reactions amongst various species in aqueous solution contained in cloud droplets. The aqueous-phase chemistry mechanism in the present study is adapted from ADOM (Acid Deposition and Oxidant Model: Venkatram et al., 1988; Fung et al., 1991). It includes mass transfer of SO2, O3, H2O2, ROOH, HNO3, NH3 and CO2 and oxidation of S(IV) to S(VI) by dissolved ozone, hydrogen peroxide, organic peroxides and oxygen (in the presence of trace metals, e.g., iron and manganese). Mass transfer between gas and aqueous phases is treated as a diffusion process, and it is written as a set of forward and backward reactions and integrated with the aqueous-phase oxidation reactions. The diffusion coefficients for the mass transfer process are determined from Fuchs and Sutugin (1971). The time integration of the chemistry system is done using a vectorized version of the Young and Boris predictor-corrector solver (Young and Boris, 1977).

The aqueous-phase chemistry is coupled with three of the explicit aerosol chemical components in AURAMS, namely sulphate, nitrate, and ammonium, whose size distribution is represented by 12 size bins equally spaced on a logarithmic scale ranging from 0.01 to 40 μ m. (Note that aerosol components in the current AURAMS also include organic aerosols and sea-salt aerosols. Although they are not directly

involved in aqueous-phase chemistry, they are however indirectly affected due to the internally mixed aerosol assumption used in AURAMS).

A bulk representation of the aqueous-phase chemistry is employed in the current AURAMS due to the computational burden for integrating the aqueous-chemistry system using the current solver (though vectorized). A bulk droplet size, determined from an average of cloud droplet size spectrum, is used for the integration, and at the end of the aqueous-phase chemistry integration the bulk mass increment of the various aerosol components is distributed across the activated size bins by using the ratios of liquid water content in each activated (or partially activated) size bin to the total (bulk) cloud water content. While it is perhaps reasonable as far as particulate sulphate is concerned, this bulk-to-size conversion is less satisfactory when it comes to other inorganic aerosol components, such as particulate nitrate and ammonium (Gong, 2002).

Finally, since a fixed bin structure is used in AURAMS, "rebinning" is required to be carried out to account for the growth of aerosols due to aqueousphase production. This is done by ensuring conservation of both mass and number (note that, while aqueous-phase chemistry will affect aerosol mass, it should not affect number concentration):

$$\begin{split} F_{i,j} &= \begin{pmatrix} m_{j,old} \\ m_{i,new} \end{pmatrix} \begin{pmatrix} \frac{m_{j+1,old} - m_{i,new}}{m_{j+1,old} - m_{j,old}} \\ \end{pmatrix} \\ F_{i,j+1} &= \begin{pmatrix} m_{j+1,old} \\ m_{i,new} \end{pmatrix} \begin{pmatrix} \frac{m_{i,new} - m_{j,old}}{m_{j+1,old} - m_{j,old}} \\ \end{pmatrix}, \end{split}$$

where $F_{i,j}$ and $F_{i,j+1}$ are partitioning factors for redistributing the "new" mass in bin i, $m_{i,new}$, after the aqueous-phase chemistry, into bins j and j+1. The sum of $F_{i,j}$ and $F_{i,j+1}$ should be unity.

2.3 Wet deposition

Wet deposition describes the processes of tracer scavenging and transport by precipitation. The scavenging process includes both tracer transfer from cloud droplets due to the autoconversion (precipitation production) process and the direct impact scavenging of aerosol particles and soluble gases by falling hydrometeors (liquid or solid). Tracers captured in precipitation will be removed from the atmosphere when the precipitation reaches the ground. However, part or all of the precipitation may evaporate before reaching the ground, in which case the tracers will remain in the atmosphere but will be vertically redistributed.

The wet deposition flux of tracer i at a given level in a vertical column can be calculated from

$$F_i(z) = (F_i(z+1) + \Delta F_i(z))(1.0 - f_{evp}(z)),$$

where f_{evp} is the fraction of precipitation loss due to evaporation. $\Delta F_i(z)$ is the input flux at level z due to either precipitation production (e.g., cloud-to-rain) or precipitation scavenging, and $F_i(z+1)$ is the flux arriving at that level from above.

The change in aqueous-phase tracer concentration in cloud droplets due to tracer transfer from cloud drops to rain drops is currently parameterized using a bulk auto-conversion (precipitation production) rate obtained from the meteorological model:

$$\frac{\partial AQ(i)}{\partial t} = -f_{ctr}AQ(i),$$

where AQ(i) is aqueous-phase species i concentration, and f_{ctr} , is precipitation production rate expressed as RWC/CWC s⁻¹.

The parameterization of impact scavenging of aerosols by precipitation is inherited from CAM (Gong et al., 2002), which is based on Slinn and Slinn (1981). The impact scavenging of soluble gases by precipitation is treated separately for irreversible scavenging and reversible scavenging. The former, applicable to species whose gas-phase concentration is much greater than their equilibrium concentration in the aqueous phase (e.g., H₂O₂, ROOH, HNO₃ and NH₃), is treated in a similar way as the scavenging of aerosols. The scavenging rate Γ is parameterized through the Sherwood number, N_{Sh} (Prupacher and Klett, 1997; Seinfeld and Pandis, 1998; Jacobson, 1999):

$$\Gamma = \frac{6N_{Sh}D_gQ_R}{D_p^2U_t}$$

and $N_{Sh} = 2 + 0.6 N_{\text{Re}}^{1/2} N_{Sc}^{1/3}$,

where D_g is the diffusivity of each gaseous species in air, Q_R is the precipitation rate, D_p is the rain-drop diameter, U_t is the terminal velocity of rain drops,

and. N_{Re} and N_{Sc} are the Reynolds and Schmidt number respectively. The terminal velocity is in turn parameterized based on Beard (1976) for three separate Reynolds number regimes: Stokes flow, with correction for slip flow condition ($D_p < 20 \ \mu m$); intermediate Reynolds numbers ($20 \ \mu m \le D_p \le 1 \ mm$); and larger drops ($1 < D_p \le 7 \ mm$).

For species whose gas-phase concentration is comparable with the equilibrium aqueous-phase concentration (e.g., SO_2 and CO_2), an equilibrium assumption is used:

$$\begin{bmatrix} HSO_{3}^{-} \end{bmatrix}_{rain} = \frac{H'_{SO2} [TSO_{2}]}{(H^{+}] + B \cdot H'_{SO2}}$$
$$\begin{bmatrix} HCO_{3}^{-} \end{bmatrix}_{rain} = \frac{H'_{CO2} [TCO_{2}]}{(H^{+}] + B \cdot H'_{CO2}}$$

where $[TSO_2]$ and $[TCO_2]$ are total SO₂ and CO₂ concentrations (gaseous + aqueous), respectively, B is a coefficient for unit conversion (between aqueousphase and gas phase concentration units), and H' is the effective equilibrium constant (combining absorption and dissociation).

Scavenging of soluble gases by snow and ice is currently only considered for HNO_3 and NH_3 (irreversible). It is parameterized as in ADOM with the scavenging rate being set to twice the rate for HSO_4 vapour, which is based on an analogy to the collection rate of supercooled water drops by snow/ice (Karamchandani et al., 1985).

3. MODEL SIMULATION AND IMPACT OF CLOUD PROCESSES

A simulation of regional oxidants and PM over eastern Canada and north-eastern U.S. using AURAMS with cloud processes has been conducted for the period of August 1 - 6, 1988. The actual simulation started from July 30, 1988, to allow for a 2-day model "spin-up". The model was run on a 71 x 71 x 28 grid with 40 km (true at 60°N) spacing in the horizontal. The 28 unevenly spaced vertical levels range from the surface to 25 km. AURAMS is currently driven by a Canadian mesoscale model (MC2) which is a flexible, fully compressible, nonhydrostatic model (Benoit et al., 1997). The evaluation of AURAMS performance for this simulation against filter and continuous chemistry (air) data collected during the EMEFS-1 period is currently underway. Here we will mainly examine the impact of cloud processes on simulated regional aerosols.

Figure 1 shows the modelled total sulfate aerosol mass concentration at the lowest model level (surface), averaged over a 24-hour period for August 1 and 4, 1988 (using GMT). The model simulation shows that high particulate sulfate up to 30 μ g m⁻³ is predicted over the eastern U.S. south of the Great





Lakes on August 1. The maximum particulate sulfate concentration then increases, reaching about 40 μ g m⁻³ (24-hour average) on August 4th with a high concentration band stretching from southeastern U.S. to southeastern Canada and the northeastern U.S. seaboard.



0 metres - Stamp: PM TOTAL S04 - Interval: 0.01 * 1.0e+01 (?]





Figure 1. Modelled total sulfate aerosol mass (μ g m⁻³) at the lowest model level, averaged over a 24-hour period: (a) August 1; (b) August 4. The top row is from the run with cloud processes; the middle row is from the run with aqueous-phase chemistry only; and the bottom row is from the run with no cloud processes. (Note that the numbers indicated on the colour scale should be multiplied by 10 to get actual values in μ g m⁻³).



Figure 2. Differences in modelled sulfate aerosol mass (x10 μ g m⁻³) on Aug. 4 (GMT) between (top panel) simulations with "all" cloud processes and without (dry); (middle panel) simulations with cloud chemistry only and without (dry); (bottom panel) simulations with "all" cloud processes and with cloud chemistry only.

Three different sensitivity runs were carried out: (1) a run with full cloud processes; (2) a run with cloud/aqueous-phase chemistry only; and (3) a run without cloud processes. The differences in modelled total sulfate aerosol mass (24-hour average) at the surface for August 4 between these runs are

presented in Figure 2. Meteorological model (MC2) predicted cloud cover (vertically integrated cloud fraction), averaged over the 24-hour period is shown in Figure 3. It is seen that the modelled sulfate aerosol mass at the surface is enhanced up to 7 μ g m⁻³ by the inclusion of aqueous-phase chemistry only, mostly (as expected) over the cloudy region. The enhancement over northern Ontario and central Quebec is significant as, most of the modelled particulate sulfate mass at the surface in this area is due to in-cloud production. The difference between the run with both cloud chemistry and wet deposition and the "dry" run reflects several processes: enhancement due to in-cloud production; reduction due to wet removal; and enhancement in the case of precipitation evaporation (injection of tracers captured in rain at the lower levels). This is better illustrated by comparing the run with "all" cloud processes and the one with chemistry only (last panel in Figure 2), where the negative areas reflect removal and the positive areas indicate the injection of tracers at lower levels due to precipitation evaporation. It is therefore interesting to see that the wet deposition process not only acts to remove tracers from the atmosphere, but also acts to redistribute tracers in the vertical, resulting in the enhancement in tracer concentration at lower levels due to evaporation of precipitation.



120 hour fcst valid 00:00Z August 04 1988

Figure 3. Meteorological-model-predicted cloud cover (vertically integrated cloud fraction) averaged over 24 hours for August 4, 1988 (GMT).

4. PRECIPITATION CHEMISTRY – COMPARISON WITH OBSERVATIONS

4.1 Observations

During the EMEFS observation period, precipitation-chemistry data were gathered by various participating sponsoring organizations and monitoring networks. These data were then combined into a common format to meet EMEFS objectives (McNaughton and Vet, 1996). Measurements commonly available from all stations include SO4=, NO3-, Cl, NH4+, Na, K, Ca, Mg, and pH, analyzed from nominally 24-hour precipitation samples. There were altogether 157 sampling sites over eastern North America involved in the study; 139 sites (some of which are co-located) are within the model domain for the current simulation. During the first week in August 1988, most of the sites had recorded some amount of precipitation, but the precipitationchemistry data were comparatively sparse (missing data due to contaminated samples, below detection limit, etc.). Nonetheless, this set of data is valuable for the evaluation of the cloud processes, particularly the wet deposition process, in the regional model.

4.2 Comparison and discussion

Comparisons presented here are based on averages over the 6-day period July 31 – August 5, 1988 (EDT). For a given measurement site the average is done for the days with available data during this period. Modelled hourly wet fluxes at the surface are extracted from the grid cells containing the measurement sites and are first averaged to obtain a daily value (according to the 24-hour sampling period). These modelled daily values are then also averaged for all the days when measurement data are available at the corresponding sites in the grid cell during the 6-day period.



Figure 4. 24-hour precipitation (mm) averaged over July 31 – August 5, 1988: modelled (on the left) and observed (on the right). (See text for the averaging procedure).

Figure 4 shows such comparison for the averaged 24-hour precipitation amount (mm). The measurements show that most of the area over eastern North America had at least 1 mm of rain during a 24-hour period on average, while the model showed much smaller amount over most of eastern U.S.. The modelled precipitation mostly corresponds to a frontal system over northern Ontario and central Quebec as indicated from the cloud cover (Figure 3), where the comparison with observation is favorable. It seems that the model is able to predict precipitation at resolved scale while having difficulty predicting sub-grid-scale convective precipitation. This is perhaps not too surprising since the sub-grid-scale precipitation is a difficult field to predict, and the model results are very sensitive to the particular convective parameterization scheme used.



(b)

Figure 5. (a) $SO4^{=}$ and (b) $NO3^{-}$ concentration in precipitation (µmol L⁻¹) averaged over July 31 – August 5, 1988, modelled (on the left) and observed (on the right).

Figure 5 shows similar comparison for the concentration of sulfate and nitrate ions in precipitation (in μ mol L⁻¹). Of all sites, the mean and standard deviation for sulfate concentration in precipitation is 42.3 and 36.8 μ mol L⁻¹, respectively from the observations, while the corresponding values from the model simulation are 74.2 μ mol L⁻¹ for the standard

deviation. The highest values from the observations are in 100 to 200 µmol L⁻¹ range, and up to 500 µmol L^{-1} from the model simulation. The high values from the model are mostly over the area where significantly lower precipitation is predicted in comparison to the observations. The high predicted concentration in precipitation can be due to a number of factors. Lower liquid water would lead to higher concentration in cloud drops (and consequently in rain drops). Another possible contributing factor is the fact that a bulk cloud-to-rain conversion rate is used for transferring tracers from cloud to rain, while in reality precipitation consists of larger drops, i.e., the higher end of the droplet spectrum, which are usually more dilute than smaller drops. The comparison for nitrate concentration in precipitation is similar but with considerably more pronounced over-prediction compared to sulfate. The average over all sites is 38.8 μ mol L⁻¹ from the observations while the average is 186.4 μ mol L⁻¹ from the model simulation. Note here that the nitrate ions in precipitation also include the direct scavenging of gaseous HNO₃ by precipitation.

Another comparison between the modelled and the observed precipitation chemistry is presented in scatter and ranked (unpaired) plots in Figure 6. Here the modelled tracer concentrations in precipitation are derived separately for the cloud-to-rain conversion portion and the precipitation-scavenging portion, in order to investigate the relative contribution due to these two pathways of wet removal. It is seen that, in the case of sulfate, the direct scavenging of aerosols by precipitation contributes rather insignificantly to the overall concentration (black dots) in precipitation. The wet deposition/removal is dominated by the cloud-to-rain enhanced reflecting the conversion. sulfate concentration in cloud due to aqueous-phase production. (It should be mentioned that possible S(IV)-to-S(VI) conversion in precipitation is not yet considered in the current model). In contrast, cloudto-rain conversion and direct precipitation scavenging contribute about equally in the case of nitrate concentration in precipitation. This is due to the fact that there is no aqueous production of nitrate in the aqueous-phase-chemistry current mechanism employed in the model; the nitrate in cloud droplets is from activation/nucleation scavenging of aerosols and condensation of gaseous HNO₃, which is comparable to the direct scavenging of aerosol and HNO₃ by precipitation. It is seen once again that the model in general over-predicts sulfate and nitrate concentration in precipitation compared to the observations.

The ranked (unpaired) plots show that the modelled and the observed sulfate in precipitation agree well up to about 50 μ mol L⁻¹. Again, the (meteorological) model is seen to seriously underpredict precipitation over this 6-day period. Finally, it needs be pointed out that although the model is seen to over-predict the tracer concentration in precipitation, the actual amount of wet removal predicted by the model is much lower than what was observed due to the significantly lower amount of precipitation in the model.



Figure 6. Model-observation comparisons of, (a) wet SO4⁼ concentration, (b) wet NO3⁻ concentration, and (c) precipitation (24-hour accumulation); scatter plot on the left and ranked (unpaired) plot on the right. In the case of (a) and (b), red denotes concentration attributed only to cloud-to-rain conversion, blue denotes concentration attributed only to precipitation scavenging, and black denotes overall.

5. SUMMARY AND CONCLUSION

In this study we have shown that, by incorporating cloud processes, the modelled regional aerosol mass concentration is significantly enhanced due to the production from aqueous-phase chemistry in cloud droplets. Wet deposition (cloud-to-rain conversion and precipitation scavenging) acts to remove tracers from the atmosphere when the precipitation reaches the ground. However, in the case of precipitation evaporation before reaching ground, the wet deposition process acts to efficiently redistribute tracers in the vertical column, which can significantly enhance the tracer concentrations at lower levels.

A preliminary comparison for a 6-day period of AURAMS model simulation with the observed precipitation-chemistry data collected during the EMEFS campaign shows that the model overpredicts ion concentrations in precipitation. The overprediction is partly due to the considerably lower amount of precipitation in the model as compared to the observed amounts. Further investigation is necessary to understand this discrepancy. The poor agreement between model and observed precipitation amounts, however, is not unexpected given the summertime convective nature. It is still very challenging to predict convective precipitation correctly in most current meteorological models. The (meteorological) model's ability to predict cloud microphysics fields and the precipitation amount will have a significant impact on the predicted precipitation chemistry and wet deposition of atmospheric tracers.

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