ALASKA ADAPTED CMAQ MODEL

Ketsiri Leelasakultum and Nicole Mölders

University of Alaska Fairbanks, Geophysical Institute, College of Natural Science and Mathematics, Department of Atmospheric Sciences, 903 Koyukuk Dr., Fairbanks, AK 99775, USA

1. INTRODUCTION

Evaluations of the Community Multiscale Air Quality (CMAQ) performance in simulating fine particulate matter ($PM_{2.5}$) and its species have been performed in many studies, and showed that the performances varies with season. Most studies found that the CMAQ performance tends to be lower in winter than summer for $PM_{2.5}$ and most species; CMAQ also is likely to underpredict sulfate (SO_4^{2-}) (Appel et al. 2008; Eder; Yu 2006; Mathur et al. 2008; Tesche et al. 2006). The reasons for the weaker performance are probably improper characterization of the weaker mixing in winter than summer, incorrect temporal emission allocations and model science processes (Tesche et al. 2006); the model bias in SO_4^{2-} that found in CMAQ v4.4 (Eder; Yu 2006) or CMAQ v4.5 (Appel et al. 2008) may be caused by excessive wet deposition (Eder; Yu 2006). Although the upgrades in CMAQ v4.7 have improved the predictions of $PM_{2.5}$ i.e. decreased the bias of SO_4^{2-} prediction through a cloud module update based on studies in the eastern US (Foley et al. 2010), the model performance still needs to be tested in different areas.

In the pristine environment of Alaska, the increased $PM_{2.5}$-concentrations in Fairbanks during winter became of major concern because they exceeded the 24h-average National Ambient Air Quality Standard (NAAQS) (Tran; Molders 2011) frequently and affected to the Fairbanks' people health (State of Alaska Epidemiology,2010). To represent the $PM_{2.5}$-concentrations in the Fairbanks non-attainment area during episodes in winter, the CMAQ model was applied. As mentioned above, the performance of CMAQ for low temperature conditions tends to be comparatively poor. Therefore some model processes in CMAQ were modified for better performance and representing of Alaska conditions.

2. DOMAIN AND INPUT DATA

The CMAQ model version 4.7.1 was applied with grid increments of 1.3km and 38 full vertical layers for a domain, which is centered over Fairbanks and covers the non-attainment area. This 1.3km domain is the innermost domain of three nested domains used in the meteorological simulations with the Weather Research and Forecasting (WRF) model version 3.1 (Skamarock et al. 2008). It is nested into a domain with 4km-grid increment covering mainly the interior of Alaska, which is again nested into a 12 km-grid increment domain covering Alaska and parts of Siberia (Fig.1). The meteorological fields were obtained by Gaudet and Staufer (2010). The emission data were gathered by Sierra Research Inc. using a bottom-up approach and generated using the Sparse Matrix Operator Kernel Emissions (SMOKE) modeling system. The inventories include emissions from home heating, airports, point sources, on-road and non-road with other area sources. The CMAQ simulation was performed for two episodes, January 23-February 10, 2008 and November 2-14, 2008 hereafter referred to as episode 1, and 2, respectively.

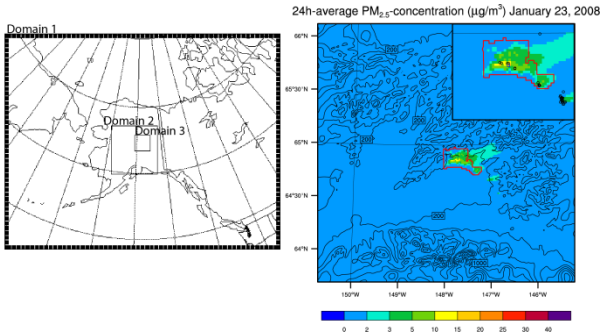

Fig. 1. Triple nested domain of WRF configuration (left, (Gaudet; Stauffer 2010)) and CMAQ simulation domain with zoom-in (domain 3, right). The red polygon, blue rectangle and black circles indicate the nonattainment area, the State Office Building (SB) and locations of point sources, respectively.

3. MODEL DEVELOPMENT

In the reference simulation, CMAQ was first run with its default configuration options. The CB05 gas-phase chemistry mechanism was used with the Euler backward interactive (EBI) solver. The AERO5 aerosol model with extensions for sea salt emissions and chemistry and a new formulation for secondary organic aerosol was used. Photolysis rates were calculated from a table of clear-sky photolysis rates that covers

domain 3. A global mass-conserving scheme (YAMO) was used for mass-conserving advection, and the K-theory eddy diffusivity scheme was used for the vertical diffusion. For cloud treatment in the model, the asymmetric convective module was used. Default initial condition (IC) and boundary condition (BC) profiles that represent clean air conditions in the eastern-half of the United States (Gipson 1999) were used.

The configuration of the Alaska adapted CMAQ model is similar to the reference simulation, except that the Sparse Matrix Vectorized GEAR (SMVGEAR) is used as chemistry solver to perform process analysis and the in-line photolysis rate module is used to consider feedback of aerosol. IC and BC profiles for Alaska conditions were first developed in accord with (Jaeschke et al. 1999; Porter 2009; Seinfeld; Pandis 2006) and measurements from the Clean Air Status and Trends Network (CASTNET). Introducing of the Alaska adapted IC and BC, $PM_{2.5}$-concentrations outside the nonattainment area are within the range of 0-2μg/m^3 (Fig.1) which is typically observed in Alaska remote areas (see IMPROVE 2011).

The lowest and highest eddy diffusivity coefficients which play an important role in vertical mixing of species during stable conditions were decreased by half and scaled based on the urban fractional land-use. Decreasing the eddy diffusivity coefficients brought about an increase of 24h-average $PM_{2.5}$-concentrations of 0.54-20.3μg/m^3 for episode 1 at the State Office Building site (SB). Note that SB site was the only observational site in the nonattainment area.

Because dispersion of air pollutants is a function of mixing height, the constant of the minimum height of the atmospheric boundary layer in the Meteorology-Chemistry Interface Processor (MCIP) was decreased from 50m to 16m in accord with the observations in Fairbanks (Wendler; Nicpon 1974). Additionally, the minimum stomatal resistance values which relate to the treatment of dry deposition gases were edited for the various land-use categories following Erisman et al. (1994). These values have been found to provide good results in simulations over Alaska with WRF/Chem (e.g. Mölders; Kramm 2010, Mölders et al. 2011, Mölders et al., 2010; 2011; Tran et al. 2011).

As CMAQ tends to underestimate sulfate under low temperature conditions, the following modifications were made to increase the simulated sulfate concentrations. The background values of Fe(III) and Mn(II), which are catalysts for the oxidation reaction of SO_2 into aqueous solutions (Seinfeld; Pandis 2006), were updated in accord with measurements in Fairbanks performed during winter 2011-2012 by Peltier (2012). Since the heterogeneous processes in clouds are the dominant path in oxidizing SO_2, the liquid-water threshold values for resolvable scale clouds were also reduced by 50% to decrease the cloud bias according to the study of Mueller et al. (2006). Finally, the parameterizations of the sulfuric acid-water nucleation rate were changed following Vehkämaki et al. (2002) who extended the formulation of the original CMAQ version code to lower temperature and wider relative humidity ranges.

4. RESULTS

4.1 Comparison of the reference with the modified model simulations

Comparison of the simulations from the reference case and the Alaska adapted CMAQ reveals that both simulations have similar temporal evolutions but the adapted CMAQ shows higher 24h-average $PM_{2.5}$-concentrations than the reference case in episode 1 at the State Office Building site (Fig. 2). The decreasing of the eddy diffusivity coefficients was the main cause for these differences. The correlation coefficient between the simulated $PM_{2.5}$-concentrations from the reference and Alaska adapted CMAQ with the observed $PM_{2.5}$-concentrations are 0.48 and 0.52, respectively.

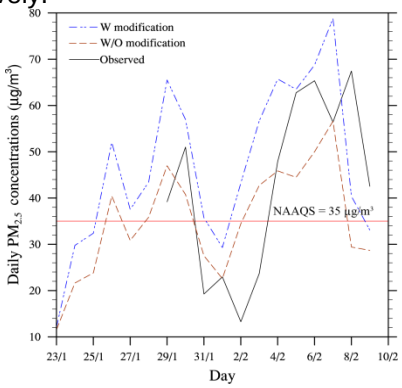

Fig. 2. Temporal evolutions of the 24h-average $PM_{2.5}$-concentrations as obtained from the reference simulations (W/O modifications, brown dashed line), the adapted CMAQ simulations (W modifications, blue dashed line) and observations (black solid line) for episode 1 at the SB site.

4.2 Evaluation of the adapted CMAQ simulated $PM_{2.5}$-concentrations

The scatter plots of the 24h-average $PM_{2.5}$-concentrations for both episodes show that simulated and observed data are agree well within a factor of two on most days. In episode 1, which had frequently high observed $PM_{2.5}$-concentrations that exceeded the NAAQS, the correlation coefficient is 0.52 (Fig. 3). In episode 2, 24h-average $PM_{2.5}$-concentrations are often lower than the NAAQS, and the correlation coefficient between simulated and observed data is lower (0.31) than for episode 1.

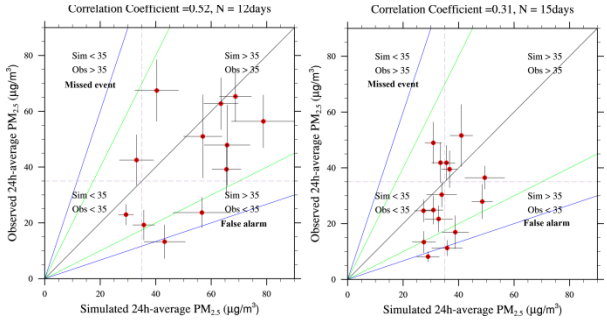

Fig.3. Scatter plots of 24h-average $PM_{2.5}$-concentrations as obtained from the adapted CMAQ during episode 1 (left) and episode 2(right) and the observations at the SB site. The green line indicates the factor of two, the blue line indicates the factor of three agreement and error bars indicate standard deviations.

Other statistics used for evaluating simulated concentrations (Cs) with observed concentrations (Co) are the root mean square error, the mean bias, the fractional bias $FB = \frac{2}{N}\sum_{i=1}^{N}\frac{C_s - C_o}{C_s + C_o} \times 100\%$, fractional error $FE = \frac{2}{N}\sum_{i=1}^{N}\left|\frac{C_s - C_o}{C_s + C_o}\right| \times 100\%$, normalized mean bias $NMB = \frac{\sum_{i=1}^{N} C_s - C_o}{\sum_{i=1}^{N} C_o} \times 100\%$, and normalized mean error $NME = \frac{\sum_{i=1}^{N}|C_s - C_o|}{\sum_{i=1}^{N} C_o} \times 100\%$. For episode 1, the means are 53.1μg/m^3 and 42.6μg/m^3 for simulated and observed 24h-average $PM_{2.5}$-concentrations, respectively. The mean bias is 10.5μg/m^3. The model simulated 10 exceedance days while only eight exceedance days were observed. The RMSE FB, FE, NMB and NME for episode 1 are 19.7μg/m^3, 22%, 35%, 25% and 39%, respectively.

For episode 2, the means of the 24h-average $PM_{2.5}$-concentrations of simulated and observed are 35.5μg/m^3 and 29.3μg/m^3, respectively. The mean bias is 6.2μg/m^3. The model simulated seven exceedance days while only six exceedance days were observed. The RMSE, FB, FE, NMB and NME for the November episode are 14.2μg/m^3, 19%, 38%, 21% and 42%, respectively. Although in episode 1, the correlation coefficient of simulations with the observation is higher than episode 2, percent bias and errors are higher as well.

Soccer plots show that the adapted CMAQ simulated four and five days outside the criteria for episode 1 and 2, respectively (Fig. 4). Three of four days in episode 1 are the days that have 24h-average $PM_{2.5}$-concentrations below the NAAQS whereas three of the five days in episode 2 are the days at the beginning of the episode, when to the model still spins up.

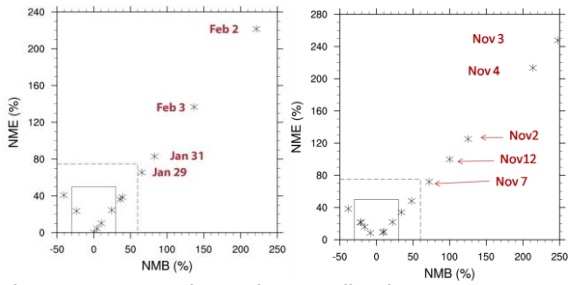

Fig. 4. Soccer plots of normalized mean errors and biases of simulated 24h-average $PM_{2.5}$-concentrations for episode 1 (left) and episode 2 (right) at the SB site.

4.3 Evaluation of the adapted CMAQ simulated sulfate concentrations

In the simulations of CMAQ that were performed without the modified sulfate treatment, the simulated composition of the 24h -average $PM_{2.5}$ aerosol overestimated the percentage of organic carbon, but strongly underestimated the percentage of sulfate and ammonium. The observations at the State Office Building suggest that sulfate makes up about 20% and 12% of the total $PM_{2.5}$ concentrations, but the CMAQ-simulations provided that sulfate makes up only 4% of the total $PM_{2.5}$-concentrations during episode 1 and 2, respectively (Fig. 5).

Potential reasons for the too low concentrations were hypothesized as the errors from the CMAQ model itself. Several changes were made to improve the sulfate simulations (see model development section). The results of the simulations with these modifications were compared with the simulations of CMAQ that has not been modified with respect to the sulfate treatment.

The introduction of the above improvements led to an increase in the percentage sulfate concentrations of total $PM_{2.5}$ at the SB site. The percentage of sulfate increased from 3.9 to 5.0%

and from 4.2 to 5.3% for episode 1 and 2, respectively.

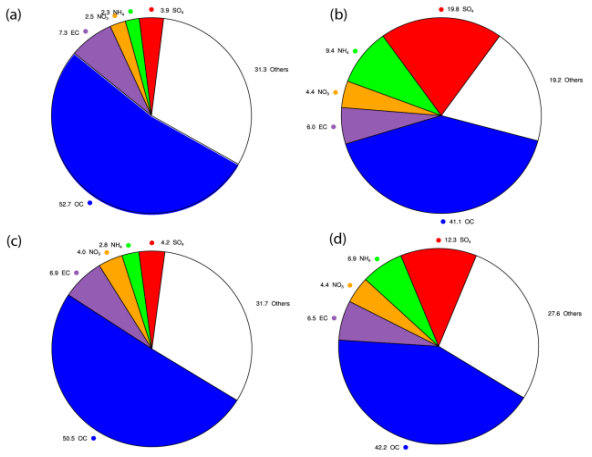

Fig. 5. Composition of (a) simulated 24h-average total $PM_{2.5}$ before introducing the modification for the sulfate treatment as obtained for episode 1, (b) observed on average over the six days for which data was available in episode 1, (c) simulated in episode 2, and (d) observed on average over the three days for which data was available in episode 2 at the SB site.

The increase of the percentage of SO_4 affects the partitioning of other species, i.e. increases the NH_4 percentage, and decreases the NO_3 and organic percentage. The enhancement of sulfur dioxide and sulfate affected the thermodynamic equilibrium of the aerosol system. The sulfate-related aerosol acidity can further be neutralized by NH_3 to form ammonium sulfate aerosol ($(NH_4)_2SO_4$) (Seinfeld; Pandis 2006). The rest of ammonia can also neutralize nitric acid (HNO_3), and form ammonium nitrate aerosol (NH_4NO_3). Introducing the changes in the parameterizations increased the mean sulfate concentration on the days, which had observed sulfate concentrations at the SB site, in the range of 1.7 to 2.1μg/m^3 and 1.6 to 2.2μg/m^3 for episode 1 and episode 2, respectively. However, these improvements still were not able to simulate sulfate concentrations as high as they were observed (Fig 6).

The statistical performance skills for sulfate specie simulated by the adapted CMAQ show a mean bias, RMSE, NMB and NME of -5.4μg/m^3, 6.1μg/m^3, -70%, 70%, respectively for episode 1. For episode 2, the mean bias, RMSE, NMB and NME are -3.1μg/m^3, 3.4 μg/m^3, -60%, 60%, respectively. The simulations from both episodes yielded high NME and NMB (exceeded 50%) which is higher than the study by Eder and Yu (2006) that had NMB and NME of -2% and 42%, respectively for other states in the US.

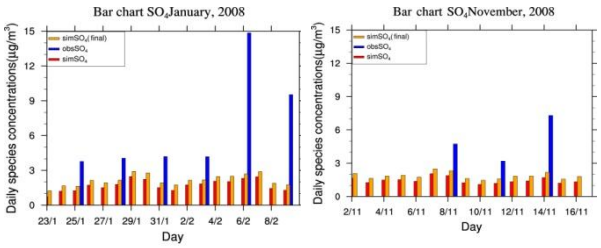

Fig. 6 Bar charts of simulated species as obtained from the adapted CMAQ without the improved sulfate treatment (red), and with the improvements for sulfate treatment (orange) and observed (blue) 24h-average SO_4concentrations for episode 1 (left) and 2 (right) at the SB site.

4.4 Process analysis of adapted CMAQ simulation

The process analysis technique was conducted to investigate the reasons of the under-prediction of sulfate at the SB site. The results show that the major contributors of sulfate specie were emissions and horizontal transport. Major removal processes for sulfate were vertical transport, and dry deposition processes, and cloud processes played a small role in sulfate formation (Fig. 7). The final modifications caused changes in the horizontal and vertical transport (Fig. 7). This means that the modifications led to changes in neighbored grid-cells that now lead to advection of modified air. On average over episode 1, the final CMAQ modification increased the contribution of sulfate from horizontal transport, cloud and aerosol processes by 0.30, 1.1×10^{-6} and 5.6×10^{-4} μg/m^3, respectively, and decreased the removal by dry deposition and vertical transport by -0.02 and 0.37μg/m^3, respectively. On average over episode 2, the final CMAQ modification increased the contribution of sulfate from horizontal transport, cloud and aerosol processes by 0.39, 8.4×10^{-7} and 4.8×10^{-4}μg/m^3, respectively, and decreased the removal by dry deposition and vertical transport by -0.02 and 0.28μg/m^3, respectively.

As emissions are the main contributor to sulfate at the SB site, we checked the emission inventory data. The comparison between the simulated SO_2-concentrations with the observed data showed that the average simulated SO_2-mixing ratio is higher than the observed SO_2-mixing ratio at the site nearby the SB site. But the percent fraction of particulate sulfate emission seems to be too low in comparison with the previous emission data that we had obtained.

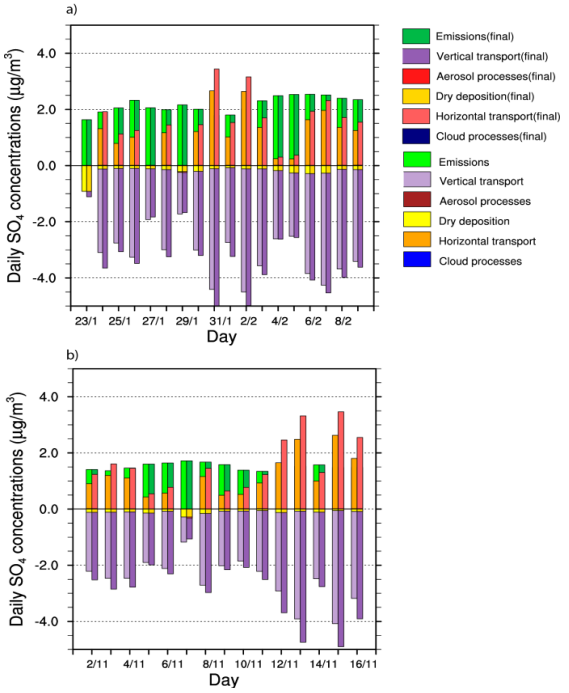

Fig. 7 Comparison of the daily contributions of individual processes to the SO_4-concentrations as simulated with CMAQ without the modified sulfate treatment and with the modified sulfate treatment at the SB site for (a) episode 1, and (b) episode 2.

5. CONCLUSIONS

We developed the Alaska adapted CMAQ model to represent the $PM_{2.5}$-concentrations in the Fairbanks nonattainment area during winter. Several changes were made to the CMAQ model code version 4.7.1 to represent Alaska conditions. Those changes include developing Alaska specific initial and background conditions, reducing the eddy diffusivity coefficients, updating the minimal stomatal resistances, and reducing the minimum atmospheric boundary layer height. Moreover, we introduced improved parameterizations for increased sulfate conditions since our simulations showed that CMAQ underestimates sulfate similar to what has been found in other regions in winter. The background values of iron and manganese were updated following the observations made in Fairbanks. The liquid-water threshold for resolvable scale cloud was decreased and the parameterization for the sulfuric acid-water nucleation was changed to be more representative for the low temperature conditions in Fairbanks.

The Alaska adapted CMAQ captures well the $PM_{2.5}$-concentrations for those days, which have 24h-average $PM_{2.5}$-concentrations that exceed the NAAQS. In the January-February episode, which has lower temperatures and higher $PM_{2.5}$-concentrations than the November episode, the model shows a correlation with the observations with a correlation coefficient of 0.52 and 35% and 39% for the NME and NMB, respectively. On the contrary for the November episode, the correlation coefficient with the observed data is 0.31 with 21% and 42% for the NME and NMB, respectively. The evaluation of the model by soccer plots indicated that most of the days that are outside criteria are days during model spin-up or the days which have $PM_{2.5}$-concentrations below the NAAQS. Therefore we suggest removing three days at the beginning of the simulations to avoid the spin-up effect.

Introduction of the improvements for sulfate treatment led to an increase in the percentage sulfate about 1% or in the range of 1.7 to 2.1µg/m^3 and 1.6 to 2.2µg/m^3 for the January-February episode and the November episode, respectively. However, the simulated sulfates are still lower than the observation data with the NME and NMB more than 50% and higher than what have observed in other US states. As the process analysis indicate that emission is the main contributor of sulfate at the State Building site, we suspect that the causes for the sulfate underestimations at this site are not only from potentially overlooked processes in the CMAQ parameterizations, but also (mainly) from the emissions. The low fraction of sulfate in primary $PM_{2.5}$-emissions in the new emission inventory should be verified.

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