Performance of CMAQ for inorganic aerosol compounds in Greater Tokyo

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1. INTRODUCTION

The air quality standard for particulate matter (PM) is set as SPM (suspended PM, 100 % cut-off at 10 μm) in Japan. The PM standard was violated at more than 40 % monitoring stations in the 1990s. In particular, the PM pollution in Greater Tokyo is very heavy, like other mega-cities in the world.

PM in Greater Tokyo is mainly composed with EC/EC and secondary inorganic species like sulfate, nitrate and ammonium. Chloride is also major in wintertime. The secondary inorganic species are semi-volatile in the atmosphere, except for sulfate. They easily change the phase between gas and aerosol, depending on temperature, relative humidity and other constituents. Therefore, they could decrease or increase with either changes in total (= gas + aerosol) amounts in the atmosphere or changes in the condition. It is required to monitor those species in the gas phase as well as the aerosol phase in order to assess their effects on the environment.

The purpose of this study is to represent concentrations of and to estimate source-apportionment for secondary inorganic species in Greater Tokyo, by using CMAQ.

2.0 MODEL DESCRIPTION

Models used in this study are MM5V3.6.1 releases in the spring of 2003, MCIP v2.2 releases in June 2003, and CMAQ v4.2.2 releases in May 2003.

Model domains are shown in Figure 1. Grid sizes are 45 km for D0, 15 km for D1 and 5 km for D2. MM5 run in D0, D1 and D2, while CMAQ run in D0 and D2. Runs in D0 were aimed to produce

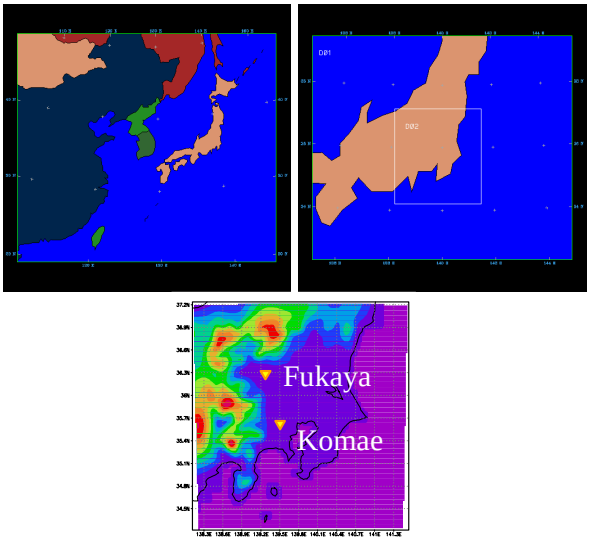

Fig. 1 Model domains: D0 (upper left) and D2 (bottom) nested in D1 (upper right). Two sites (Fukaya and Komae) are also shown in D2.

initial and boundary conditions for CMAQ runs in D2. Air columns up to 100 hPa were sliced into 35 layers in MM5 and 16 layers in CMAQ. The lowest model height was ~25 m in MM5 and CMAQ.

MM5 run with no interaction between D0 and D1/2 but with two-way nesting between D1 and D2. Objectively analyzed meteorological fields by the Japanese Meteorological Agency (RANAL, 20 km resolution at 00Z and 12Z) were used to drive MM5 in D0 and D1. Physical options of MM5 in D1/2 were Grell cumulus scheme, MRF boundary layer parameterization, simple ice, no shallow, and multi-layer soil model.

CMAQ was configured with CB4-AQ-AE3 by MEBI, PPM, EDDY and RADM-cloud.

Emission inventories used are the 1 x 1 deg. emissions prepared for ACESS (ACE-Asia and Trace-P Modeling and Emission Support System) by Streets et al. (2003) for D0 and ca. 5 x 5 km emissions produced by National Institute of Environmental Studies of Japan and Japan Clean Air Program.

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3.0 MODEL PERFORMANCE

3.1 Field Measurements

Many field campaigns have been done to understand the behavior of PM in Greater Tokyo. Unfortunately, most of them did not bring useful datasets with high-PM episodes. We chose two campaigns for model validation; December 9 to 10, 1999 as a winter case and July 31 to August 1, 2001 as a summer case. To those two-day episodes, pre-calculations of MM5 and CMAQ were performed for five days in D0 and one day in D1/2.

3.2 Winter Case

Figure 2 compares time series of measurements of gases (NO_2, SO_2 and O_3) and aerosol components at Fukaya (see Figure 1) with estimates by CMAQ. General variations of NO_2 and O3 are well produced by CMAQ. However, high concentrations are underestimated by CMAQ. SO_2 are consistently and considerably overestimated. In this winter case, measurements of NMHC composition are available, but higher concentrations are not reproduced by CMAQ (not shown). The measured peak concentrations of nitrate are well agreed by the estimated but predicted earlier. Levels of ammonium are comparative between measurement and calculation. However, sulfate is mostly doubled. Similar results were obtained for other measurement sites.

3.3 Summer Case

Figure 3 shows measurements and estimates of gases and aerosol components at Komae (see Figure 1). In this summer case, also compared are measurements of nitric acid and ammonia in the gas phase collected with the annular-denuder and filter-pack sampling system. In this case, CMAQ overestimates NO_2 but underestimate O_3 at higher concentrations. The time series of SO_2 is mostly traced by CMAQ. Measurements of particulate and gaseous nitrate are well predicted by CMAQ, except for the spike of particulate nitrate in the evening of August 1. Although concentration levels of particulate ammonium mostly agree between the observation and calculation, but CMAQ considerably overestimates the gaseous ammonia measurements. In these two days, sulfate concentrations did not change so much. However, calculated sulfate largely varies in August 1. Results at another site near Fukaya are not so good as those at Komae.

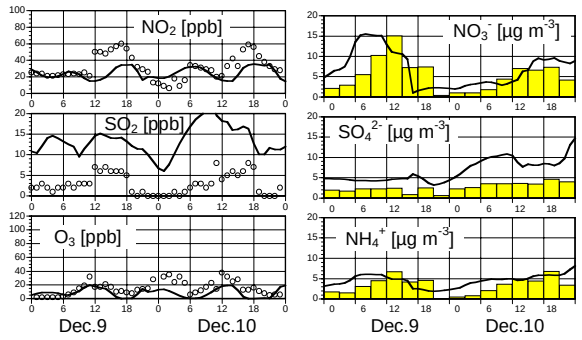

Fig. 2 Measured (circles and columns) and Calculated (bold lines) concentrations at Fukaya

4.0 SUMMARY

CMAQ was applied to Greater Tokyo for two field campaigns; one in winter and another in summer. General features of variation in gaseous and aerosol components were well reproduced by CMAQ. However, large errors were found for certain species, sites and days. Some of them seem to be related with the reality of meteorological fields produced by MM5. For species, we are thinking that SO_2 and sulfate must be important, because sulfate remarkably affects the gas-aerosol partitioning of nitrate and ammonium.

REFERENCE

Streets, D. et al. (2003) An inventory of gaseous and primary aerosol emissions in Asia in the year 2000, J. Geophys. Res., In press.

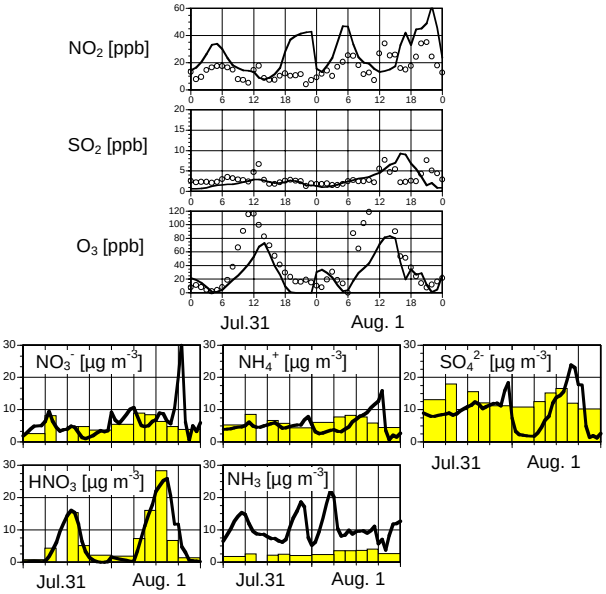

Fig. 3 Measured (circles and columns) and Calculated (bold lines) concentrations at Komae