APPLICATION OF KOREAN AIR QUALITY MODELING SYSTEM NAMED GMAF TO WINTER AND SPRING IN 2018

HyeonYeong Park* and SeogYeon Cho
Department of Environmental Engineering, Inha University, Incheon, 22212, Republic of Korea

1. INTRODUCTION

The global to mesoscale air quality forecast and analysis system (GMAF) linking a mesoscale to a global model system was developed by Cho et al. (2021). The GMAF system was used to predict air pollutants concentrations and investigate their production mechanisms in Korea. In the previous studies using the GMAF (Cho et al., 2021; Lim et al., 2022; Park et al., 2023), the researchers focused on atmospheric phenomena in 2016 and not severe $PM_{2.5}$ pollution period. The $PM_{2.5}$ pollution frequently occurred from winter to spring in 2018 and 2019 in Korea but the GMAF was not used to predict atmospheric phenomena during those periods.

In this study, we introduced again the Korean air quality system named GMAF and used it to predict $PM_{2.5}$, SO_2, NO_2, O_3 in Seoul, capital of Korea, from winter to spring in 2018. Also, we assessed model performance using three statistical indicators: correlation coefficient (R), normalized mean bias (NMB), normalized mean error (NME).

2. METHODS AND MATIRIALS

2.1 Description of GMAF

The global to mesoscale air quality forecast and analysis system (GMAF) was developed by Cho et al. (2021). Figure 1 shows the schematic diagram of GMAF framework. The GMAF uses the Weather Research and Forecasting (WRF) model version 3.6 as an atmospheric model and the Community Multi-scale Air Quality model (CMAQ) version 5.3.1 as an air quality model to predict concentrations of air pollutants. The CMAQ was modified to optimize its performance for Korea. The modification of CMAQ was described in section 2.3.

**Corresponding author:* HyeonYeong Park, Department of Environmental Engineering, Inha University, Incheon, 22212, Republic of Korea, e-mail: hypark@inha.edu

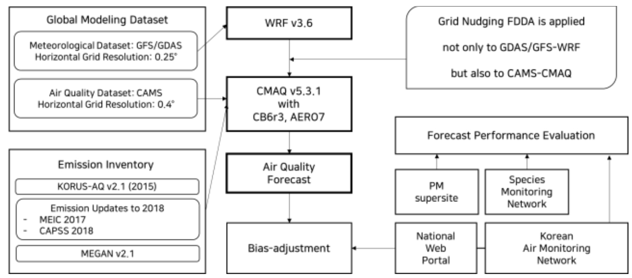

Figure 1. Schematic diagram of GMAF

2.2 Grid Nudging and CAMS dataset

The most important characteristic of GMAF is to apply the grid nudging based on four-dimensional data assimilation (FDDA) not only to the WRF but also to the CMAQ with a global data assimilation system. For application of the grid nudging based on FDDA to the CMAQ and WRF, the Copernicus Atmosphere Monitoring Service (CAMS) forecast and reanalysis dataset created by the European centre for medium-range weather forecasts (ECMWF) with 0.4° grid resolution and the Global Data Assimilation System (GDAS) dataset with 0.25° grid resolution were used, respectively. The formula of grid nudging based on FDDA was defined in equation 1.

$$\frac{dY_{model}}{dt} = F(x, Y_{model}) + W(x)(Y_{obs} - Y_{model}) \quad (1)$$

where Y is dependent variable, x is independent spatial variable, F denotes discretized form of the governing equation, and W is nudging coefficient. The dependent variable Y is calculated by equation 2 and 3 for the WRF and CMAQ.

$$Y = \alpha(P_{top} - P_{surface}) \quad \text{for the WRF} \quad (2)$$
$$Y = \varphi J_s \quad \text{for the CMAQ} \quad (3)$$

where α is meteorological variable of the WRF and P_{top} and $P_{surface}$ mean air pressure at the top and surface of the domain, respectively. The φ is mass concentration of chemical species and J_s denotes vertical Jacobian of the terrain-influenced coordinates. The nudged variables and their nudging coefficients can be found in Cho et al. (2021).

The CAMS dataset uses a hybrid and sigma-pressure vertical coordinate with 137 levels and the top level is 0.01 hPa. It provided concentrations of 14 gaseous species and seven aerosol species with 3-hour intervals. The gaseous chemical species provided from CAMS were nitrogen oxide (NO), nitrogen dioxide (NO_2), ozone (O_3), sulfur dioxide (SO_2), formaldehyde (HCHO), hydrogen peroxide (H_2O_2), nitric acid (HNO_3), hydroxyl radical (OH), peroxyacetyl nitrates (PAN), carbon monoxide (CO), methane, ethane, and propane. The aerosol species were dust and sea-salt with three size bins, hydrophobic black carbon, hydrophilic black carbon, hydrophobic organic matter, hydrophilic organic matter, and sulfate. Nitrate and ammonium were included as aerosol species of CAMS dataset from July 9 in 2019. Among chemical species of CAMS, SO_2, CO, NO, NO_2, isoprene, O_3, dust, sea-salt were used as nudged variables. Cho et al. (2021) applied nudging coefficient depending on outer domain and inner domain of nested domain system. Since we used uniform grid system (Figure 3) in this work, we interpolated the CAMS dataset to the model domain except South Korea.

2.3 Modification of CMAQ

To improve the model performance, a few mechanisms in CMAQ were modified. First, we modified the wet scavenging mechanism to suppress overestimation of wet deposition, second, modifying the vertical mixing height, third, uptake coefficient of N_2O_5, and fourth, scale factor of potential volatile organic compounds from combustion (pcVOC) emission.

2.3.1 Below-cloud Scavenging

Current CMAQ assumes that accumulation and coarse modes particles are completely absorbed into cloud and rainwater. This assumption is not valid in the below-cloud area and may cause overestimation amount of wet scavenging, subsequently underestimation of particulate matter concentration in the below-cloud area. Thus, Cho et al. (2021) modified the below-cloud scavenging rate by equation 4.

$$\frac{\partial c(t)}{\partial t} = -\Lambda c(t) \qquad (4)$$

The c(t) means the concentration of particulate chemical species at time ‘t’, and the Λ is the scavenging coefficient. The below-cloud scavenging coefficient was calculated by semiempirical formula from Slinn (1983) with collection efficiency depending on aerosol and droplet diameter.

2.3.2 Vertical Mixing Height

CMAQ’s vertical diffusion calculation requires a vertical eddy diffusivity (K_z) and set the minimum value of K_z ($K_{z,min}$) defined as equation 5 to avoid too low vertical diffusion.

$$K_{z,min} = K_{ZL} + (K_{ZU} - K_{ZL}) \times F_{urban} \qquad (5)$$

where K_{ZU} and K_{ZL} mean highest K_z and lowest K_z. CMAQ version 4 set K_{ZU} and K_{ZL} equal to 2.0 and 0.5. However, the values of K_{ZU} and K_{ZL} were changed to 1.0 and 0.01 in CMAQ version 5, which cause suppression of vertical diffusion and subsequently overestimation of air pollutants concentration at the surface. Thus, we adapted the values of K_{ZU} and K_{ZL} of CMAQ version 4 in this work.

2.3.3 Uptake Coefficient of N_2O_5

The particulate nitrate is formed by thermodynamic equilibrium between gaseous NH_3 and HNO_3. The gaseous HNO_3 is mainly produced by NO_2+OH reaction during the daytime, and heterogeneous N_2O_5 hydrolysis during the nighttime. Due to uncertainty of uptake coefficient of N_2O_5 on the aerosol surface, CMAQ provides various options for estimation of the uptake coefficient of N_2O_5. Davis et al. (2008) and Bertram and Thorton (2009)’s N_2O_5 uptake coefficient formulas are provided as default option to estimate uptake coefficient of N_2O_5 on accumulation and coarse mode particles.

Park and Cho (2020) found that nighttime HNO_3 formation using N_2O_5 uptake coefficient calculated by Davis et al. (2008)’s formula led to overestimation of nitrate concentration in Korea. Therefore, we considered organic coating inhibition on the aerosol surface (Anttila et al., 2006) to suppress overestimate uptake of N_2O_5 and revised the formula of N_2O_5 uptake coefficient as equation 6.

$$\frac{1}{\gamma_{N2O5}} = \frac{1}{\gamma_{N2O5,core}} + \frac{1}{\gamma_{N2O5,coating}} \qquad (6)$$

We calculated the ‘$\gamma_{N2O5,core}$’ and ‘$\gamma_{N2O5,coating}$’ by using Davis et al. (2008)’s formula and equation 7.

$$\gamma_{N2O5,coating} = \frac{4R_{gas}TH_{org}D_{org}R_c}{c_{N2O5}\ell R_p} \qquad (7)$$

H_{org} and D_{org} denote the Henry’s constant and the molecular diffusion coefficient of N_2O_5 in the organic coating, respectively. And R_c and R_p mean the radius of the aerosol core and that of the total aerosol, respectively. ℓ is the coating thickness and C_{N2O5} is the average velocity of N_2O_5 in the gaseous phase.

2.3.4 Parameter of pcVOC Emission

The potential secondary organic aerosol from combustion (pcSOA) is a new SOA surrogate species in AERO7. Cho et al. (2021) figured out that the pcSOA concentration was overestimated and subsequently organic matter (OM) concentration was overpredicted in Korea. The pcSOA is formed by an OH oxidation of precursor named pcVOC. The pcVOC emission is calculated by multiplying scale factor to POA emission without theoretical way. Thus, Park et al. (2023) revised pcVOC emission scale factor equal to 0.0155 mole-pcVOC/g-POA based on sensitivity simulations focused on Seoul conducted by Cho et al. (2021). In this study, we also used the same pcVOC emission scale factor used in Park et al. (2023).

2.4 Study Area and Model Configuration

Seoul is the capital of South Korea and most populous city. Seoul is located in northwestern area of South Korea as shown in Figure 2 and frequently suffered from $PM_{2.5}$ pollution in winter and spring. Therefore, we selected Seoul as the study area of interest in this study.

Figure 2. Location of Seoul (star mark).

As described above, the WRF version 3.6 and the modified CMAQ version 5.3.1 were used to predict air pollutants concentrations. The 3rd release of carbon bond version 6 (CB6r3) and AERO7 were used as gas-phase chemistry mechanism and aerosol module of CMAQ, respectively. As shown in Figure 1, the Clean Air Policy Support System (CAPSS) emission 2018 and the Multi-resolution Emission Inventory in China (MEIC, Li et al., 2017; Zheng et al., 2018) 2017 were used as anthropogenic emission inventory for South Korea and China, respectively. The Korea-United Staes Air Quality study (KORUS-AQ) anthropogenic emission inventory version 2.1 was also used for the rest of region. And the natural emission was generated by the Model of Emissions of Gases and Aerosols from Nature (MEGAN) version 2.1 (Guenther et al., 2012).

Figure 3 shows the model domain used in this study. It consists of 391 rows and 288 columns with 12 km grid spacing. The model domain includes Korean peninsula, China, Japan, and parts of Russia.

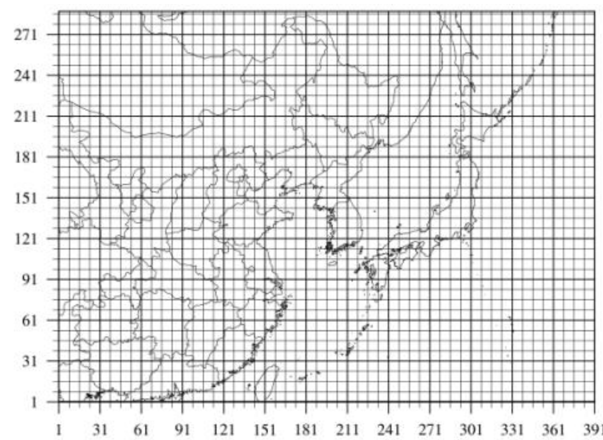

Figure 3. Model domain of GMAF

The model simulation was conducted with spin-up of 10 days. The study period of interest is from January 1st to March 31th in 2018, where severe $PM_{2.5}$ episodes were occurred in Seoul.

2.5 Evaluation of Model Performance

Emery et al. (2017) suggested benchmarks of forecasting air pollutants using chemical transport model. We used three statistical indicators, correlation coefficient (R), normalized mean bias (NMB), and normalized mean error (NME) to evaluate the model performance. Those three statistical indicators were defined in equation 8, 9, and 10.

$$R = \frac{\sum_{i=1}^{i=N}(O_i-\bar{O})(M_i-\bar{M})}{\sqrt{\sum_{i=1}^{i=N}(O_i-\bar{O})^2}\sqrt{\sum_{i=1}^{i=N}(M_i-\bar{M})^2}} \quad (8)$$

$$NMB\ (\%) = \frac{\bar{M}-\bar{O}}{\bar{O}} \times 100 \quad (9)$$

$$NME\ (\%) = \frac{1}{N}\frac{\sum_{i=1}^{i=N}|M_i-O_i|}{\bar{O}} \quad (10)$$

The model performance was evaluated by comparing modeled $PM_{2.5}$, SO_2, NO_2, and O_3 concentrations to observed ones which were monitored in monitoring station located in Seoul.

3. RESULTS

Table 1 to Table 4 summarize observed and modeled $PM_{2.5}$, SO_2, NO_2, and O_3 concentrations, and model performances. The model showed reasonable performance in $PM_{2.5}$, NO_2, and O_3 predictions.

The Rs of $PM_{2.5}$ prediction were around 0.9, which satisfy the goal of R for $PM_{2.5}$ prediction (>0.7) by Emery et al. (2017). Also, the NMBs and NMEs satisfied the goal of them (<±10% for NMB, <35% for NME).

The Rs of NO_2 prediction ranged from 0.75 to 0.91. The NMBs of NO_2 prediction were ±2% in January and February 2018 and it increased to 20% in March 2018. The NMEs of NO_2 prediction were 18~34%.

Although, the Rs of O_3 prediction were relatively lower than those of $PM_{2.5}$ and NO_2 predictions, they satisfied the goal of R for O_3 prediction (>0.75) in January, March, and total period. The modeled O_3 was underestimated with the NMBs from -8% to -17%. The NMBs of O_3 prediction satisfied the criteria but the NMEs of O_3 prediction did not satisfy even the criteria (<± 15% for NMB, <25% for NME).

The model highly underestimated SO_2 concentration during all study period (NMBs around -50%). Subsequently, the Rs and NMEs of SO_2 prediction were worse than those of $PM_{2.5}$, NO_2, and O_3 predictions. Since the detection limit of the SO_2 monitoring equipment was so high that low SO_2 concentration could not captured. Therefore, the observed SO_2 concentrations were 1~3 ppb higher in the temporal variations (not shown).

Table 1. Monthly average of observed and modeled $PM_{2.5}$ concentrations in Seoul from January to March, and model performances.

	Avg. Obs. μg/m³	Avg. Model μg/m³	R	NMB, %	NME, %
January	32.2	32.1	0.96	-0.1	20
February	30.2	27.5	0.90	-9	20
March	34.1	30.1	0.88	-12	28
Total	32.3	30.0	0.88	-7	23

Table 2. Monthly average of observed and modeled SO_2 concentrations in Seoul from January to March, and model performances.

	Avg. Obs. ppb	Avg. Model ppb	R	NMB, %	NME, %
January	5.5	2.6	0.56	-53	53
February	5.5	2.6	0.61	-52	54
March	4.7	2.3	0.69	-51	54
Total	5.2	2.5	0.63	-52	54

Table 3. Monthly average of observed and modeled NO_2 concentrations in Seoul from January to March, and model performances.

	Avg. Obs. ppb	Avg. Model ppb	R	NMB, %	NME, %
January	35.0	35.8	0.91	2	18
February	34.1	33.3	0.83	-2	22
March	33.1	39.9	0.75	20	34
Total	34.1	36.4	0.82	7	24

Table 4. Monthly average of observed and modeled O_3 concentrations in Seoul from January to March, and model performances.

	Avg. Obs. ppb	Avg. Model ppb	R	NMB, %	NME, %
January	13.7	11.9	0.85	-13	29
February	18.3	16.8	0.70	-8	34
March	25.8	21.5	0.79	-17	31
Total	19.3	16.7	0.80	-13	32

4. CONCLUSION

In this study, we conducted a model simulation for Seoul in Korea using GMAF developed by Cho et al. (2021) from January to March 2018. In GMAF, the grid nudging based on FDDA was applied not only to the WRF but also to the CMAQ. Also, below-cloud wet scavenging, vertical mixing height, N_2O_5 uptake coefficient, and pcVOC emission scale factor in CMAQ were modified. The model performance for predicting $PM_{2.5}$, SO_2, NO_2, and O_3 was assessed by R, NMB, and NME. The model showed acceptable agreement with observation in predictions of $PM_{2.5}$, NO_2, and O_3 with satisfaction of R, NMB, and NME's goal and criteria.

Reference

Anttila, T., Kiendler-Scharr, A., Tillmann, R. and Mentel, T.F., 2006. On the reactive uptake of gaseous compounds by organic-coated aqueous aerosols: Theoretical analysis and application to the heterogeneous hydrolysis of N2O5. *The Journal of Physical Chemistry A*, *110*(35), pp.10435-10443.

Bertram, T.H. and Thornton, J.A., 2009. Toward a general parameterization of N2O5 reactivity on aqueous particles: the competing effects of particle liquid water, nitrate and chloride. *Atmospheric Chemistry and Physics*, *9*(21), pp.8351-8363.

Cho, S., Park, H., Son, J. and Chang, L., 2021. Development of the global to mesoscale Air quality forecast and analysis system (GMAF) and its application to PM2.5 forecast in Korea. *Atmosphere*, *12*(3), p.411.

Davis, J., Bhave, P., Foley, K., 2008. Parameterization of N2O5 reaction probabilities on the surface of particles containing ammonium, sulfate, and nitrate. *Atmospheric Chemistry and Physics,* 8(17), 5295-5311.

Emery, C., Liu, Z., Russell, A.G., Odman, M.T., Yarwood, G. and Kumar, N., 2017. Recommendations on statistics and benchmarks to assess photochemical model performance. *Journal of the Air & Waste Management Association*, 67(5), pp.582-598.

Guenther, A., Jiang, X., Heald, C.L., Sakulyanontvittaya, T., Duhl, T.a., Emmons, L., Wang, X., 2012. The Model of Emissions of Gases and Aerosols from Nature version 2.1 (MEGAN2.1): an extended and updated framework for modeling biogenic emissions. *Geoscientific Model Development,* 5(6), 1471-1492.

Li, M., Liu, H., Geng, G., Hong, C., Liu, F., Song, Y., Tong, D., Zheng, B., Cui, H., Man, H., 2017. Anthropogenic emission inventories in China: a review. *National Science Review*, 4(6), 834-866.

Lim, J., Park, H. and Cho, S., 2022. Evaluation of the ammonia emission sensitivity of secondary inorganic aerosol concentrations measured by the national reference method. *Atmospheric Environment*, *270*, p.118903.

Park, H.Y., Hong, S.C., Lee, J.B. and Cho, S.Y., 2023. Modeling of Organic Aerosol in Seoul Using CMAQ with AERO7. *Atmosphere*, *14*(5), p.874.

Park, H.Y. and Cho, S.Y., 2020, The effects of NH3 emission reduction on secondary inorganic aerosols evaluated by CMAQ, *Journal of Korean Society for Atmospheric Environment*, 36(3), 375-387.

Slinn, W., 1983. Precipitation scavenging, in atmospheric sciences and power production–1979. Division of Biomedical Environmental Research, US Department of Energy, Washington, DC, 466-532.

Zheng, B., Tong, D., Li, M., Liu, F., Hong, C., Geng, G., Li, H., Li, X., Peng, L., Qi, J., 2018. Trends in China's anthropogenic emissions since 2010 as the consequence of clean air actions. *Atmospheric Chemistry and Physics,* 18(19), 14095-14111.