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Development of Korean Air Quality Prediction System version 1 (KAQPS v1): an operational air quality prediction system with focuses on practical issues

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Abstract

For the purpose of providing reliable and robust air quality predictions, an operational air quality prediction system was developed for the main air quality criteria species in South Korea (PM_{10}, $PM_{2.5}$, CO, O_3, and SO_2). The main caveat of the system is to prepare the initial conditions (ICs) of the Community Multi-scale Air Quality (CMAQ) model simulations using observations from the Geostationary Ocean Color Imager (GOCI) and ground-based monitoring networks in northeast Asia. The performance of the air quality prediction system was evaluated during the Korea-United States Air Quality Study (KORUS-AQ) campaign period (1 May–12 June 2016). Data assimilation (DA) of optimal interpolation (OI) with Kalman filter was used in this study. One major advantage of the system is that it can predict not only particulate matter (PM) concentrations but also PM chemical composition including five main constituents: sulfate (SO_4^{2-}), nitrate (NO_3^-), ammonium (NH_4^+), organic aerosols (OAs), and elemental carbon (EC). In addition, it is also capable of predicting the concentrations of gaseous pollutants (CO, O_3 and SO_2). In this sense, this new operational air quality prediction system is comprehensive. The results with the ICs (DA RUN) were compared with those of the CMAQ simulations without ICs (BASE RUN). For almost all of the species, the application of ICs led to improved performance in terms of correlation, errors, and biases over the entire campaign period. The DA RUN agreed reasonably well with the observations for PM_{10} (IOA = 0.60; MB = -13.54) and $PM_{2.5}$ (IOA = 0.71; MB = -2.43) as compared to the BASE RUN for PM_{10} (IOA = 0.51; MB = -27.18) and $PM_{2.5}$ (IOA = 0.67; MB = -9.9). A significant improvement was also found with the DA RUN in terms of bias. For example, for CO, the MB of -0.27 (BASE RUN) was greatly enhanced to -0.036 (DA RUN). In the cases of O_3 and SO_2, the DA RUN also showed better

performance than the BASE RUN. Further, several more practical issues frequently encountered in the operational air quality prediction system were also discussed. In order to attain more accurate ozone predictions, the DA of NO_2 mixing ratios should be implemented with careful consideration of the measurement artifacts (i.e., inclusion of alkyl nitrates, HNO_3, and PANs in the ground-observed NO_2 mixing ratios). It was also discussed that, in order to ensure accurate nocturnal predictions of the concentrations of the ambient species, accurate predictions of the mixing layer heights (MLH) should be achieved from the meteorological modeling. Several advantages of the current air quality prediction system, such as its non-static free parameter scheme, dust episode prediction, and possible multiple implementations of DA prior to actual predictions, were also discussed. These configurations are all possible because the current DA system is not computationally expensive. In the ongoing and future works, more advanced DA techniques such as the three-dimensional variational (3DVAR) method and ensemble Kalman filter (EnK) are being tested and will be introduced to the Korean operational air quality forecasting system.

Keywords: Air quality prediction; Particulate matter (PM); Geostationary satellite sensor (GOCI); Air Korea; Data assimilation (DA); Dust episode predictions; NO_2 measurement artifacts

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1. Introduction

Air quality has long been considered an important issue in climate change, visibility, and public health, and it is strongly dependent upon meteorological conditions, emissions, and the transport of air pollutants. Air pollutants typically consist of atmospheric particles and gases such as particulate matter (PM), carbon monoxide (CO), ozone (O_3), nitrogen dioxide (NO_2), and sulfur dioxide (SO_2). These aerosols and gases play important roles in anthropogenic climate forcing both directly (Bellouin et al., 2005; Carmichael et al., 2009; IPCC, 2013; Scott et al., 2014) and indirectly (Bréon et al., 2002; IPCC, 2013; Penner et al., 2004; Scott et al., 2014) in influencing the global radiation budget. Among the various air pollutants, PM and surface O_3 are the most notorious health threats, as has been stated by several previous studies (e.g. Carmichael et al., 2009; Dehghani et al., 2017; Khaniabadi et al., 2017).

With the stated importance of atmospheric aerosols and gases, considerable research efforts have been made to monitor and quantify their amounts in the atmosphere through satellite-, airborne-, and ground-based observations as well as chemistry-transport model (CTM) simulations. In South Korea, the Korean Ministry of the Environment (KMoE) provides real-time chemical concentrations as measured by ground-based observations for six criteria air pollutants (PM_{10}, $PM_{2.5}$, O_3, CO, SO_2, and NO_2) at the Air Korea website (https://www.airkorea.or.kr). In addition, the National Institute of Environmental Research (NIER) of South Korea provides air quality (chemical weather) predictions using multiple CTM simulations. Air quality predictions are another crucial element for protecting public health through the forecasting of high air pollution episodes in advance and alerting citizens about these high episodes. In this context, reliable and robust chemical weather forecasts are

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necessary to avoid any confusion caused by poor predictions given by CTM simulations.

Although there are various datasets representing air quality, limitations remain in the observations and model outputs. Specifically, observation data are, in general, known to be more accurate than model outputs, but they have spatial and temporal limitations. Unlike observation data, models can provide meteorological and chemical information without any spatial and temporal data discontinuity, but they do have an issue of inaccuracy. The major causes of uncertainty in the results of CTM simulations are introduced from imperfect emissions, meteorological fields, initial conditions (ICs), and physical and chemical parameterizations in the models (Carmichael et al., 2008). In order to minimize the limitations and maximize the advantages of observation data and model outputs, there have been numerous attempts to provide accurate and spatially- as well as temporally- continuous information on chemical composition in the atmosphere by integrating observation data with model outputs via data assimilation (DA) techniques.

Although the Korean operational numerical weather prediction (NWP) carried out by the Korea Meteorological Administration (KMA) employs various DA techniques, almost no previous efforts have been made to develop a chemical weather prediction system with DA in South Korea. Therefore, in the present study, an operational chemical weather prediction system named as Korean Air Quality Prediction System version 1 (KAQPS v1) was developed by preparing ICs via DA for the Community Multi-scale Air Quality (CMAQ) model (Byun and Schere, 2006; Byun and Ching, 1999) using satellite- and ground-based observations for particulate matter (PM) and atmospheric gases such as CO, O_3, and SO_2. The performances of the system were then demonstrated during the period of the Korea-United States Air Quality Study (KORUS-AQ) campaign (1 May – 12 June 2016) in South Korea.

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In this study, the optimal interpolation (OI) method with the Kalman filter was applied in order to develop an operational air quality prediction system, since this method is still useful and viable in terms of computational cost and performance. The performance of the method is almost comparable to that of the three-dimensional variational (3DVAR) method, as shown in Tang et al. (2017). More complex and advanced DA techniques are currently being and will continue to be applied to current air quality prediction systems. These works are now in progress.

In addition, this manuscript also discusses several practical issues frequently encountered in the operational air quality predictions such as: i) DA of NO_2 mixing ratios for accurate ozone prediction with a careful consideration of measurement artifacts; ii) the issue of the nocturnal mixing layer height (MLH) for nocturnal predictions; iii) predictions of dust episodes; iv) the use of non-static free parameters; and v) the influences of multiple implementations of the DA before the actual predictions.

The details of the datasets and methodology used in this study are described in Sect. 2. The results of the developed operational chemical weather prediction system are discussed in Sect. 3, and then a summary and conclusions are given in Sect. 4.

2. Methodology

The operational air quality prediction system was developed using the CMAQ model along with meteorological inputs provided by the Weather Research and Forecasting (WRF) model (Skamarock et al., 2008). The ICs for the CMAQ model simulations were prepared via the DA method using satellite-retrieved and ground-based observations. The performances of the developed prediction system were evaluated using ground in-situ data. The models, data, and DA technique are described in detail in the following sections.

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2.1 Meteorological and chemistry-transport modeling

2.1.1 WRF model simulations

The WRF model has been developed for providing mesoscale numerical weather prediction (NWP). It has also been used to provide meteorological input fields for CTM simulations (Appel et al., 2010; Chemel et al., 2010; Foley et al., 2010; Lee et al., 2016; Park et al., 2014). In this study, WRF v3.8.1 with the Advanced Research WRF (ARW) dynamic core was applied to prepare the meteorological inputs for the CMAQ model simulations. The National Centers for Environmental Prediction Final Analysis data (NCEP FNL) were chosen for the ICs and boundary conditions (BCs) for the WRF simulations. In order to minimize meteorological field error, the objective analysis (OBSGRID) nudging was conducted using the NCEP Automated Data Processing (ADP) global upper-air/surface observational weather data. The model domain for the WRF simulations covers Northeast Asia with a horizontal resolution of 15×15 km^2, having a total of 223 latitudinal and 292 longitudinal grid cells. The size of the WRF domain is slightly larger than that of the CMAQ domain, as shown in Fig. 1. The meteorological data also have 27 vertical layers from the surface (1000 hPa) to 50 hPa.

2.1.2 CMAQ model simulations

The CMAQ v5.1 model was used to estimate the concentrations of the atmospheric chemical species over the domain, as shown in Fig. 1. The CMAQ domain has 204 latitudinal and 273 longitudinal grid cells in total, and also has a 15×15 km^2 horizontal resolution and 27 sigma vertical layers. For anthropogenic emissions, KORUS v1.0 emissions (Woo et al.,

2012) were used. The KORUS v1.0 emissions cover almost all of Asia, and are based on three emission inventories: the Comprehensive Regional Emissions inventory for Atmospheric Transport Experiment (CREATE) for East Asia excluding Japan; the Model Inter-Comparison Study for Asia (MICS-Asia) for Japan; and the Studies of Emissions and Atmospheric Composition, Clouds and Climate Coupling by Regional Surveys (SEAC4RS) for South and Southeast Asia.

Biogenic emissions were prepared by running the Model of Emissions of Gases and Aerosols from Nature (MEGAN v2.1; Guenther et al., 2006, 2012) with a grid size identical to that of the CMAQ model simulations. For the MEGAN simulations, the MODIS land cover data (Friedl et al., 2010) and improved leaf area index (LAI) based on MODIS datasets (Yuan et al., 2011) were utilized. Pyrogenic emissions were obtained from the Fire Inventory from NCAR (FINN; Wiedinmyer et al., 2006, 2011). The lateral BCs for the CMAQ model simulations were prepared using the global model results of the Model for Ozone and Related chemical Tracers version 4 (MOZART-4; Emmons et al., 2010) at every 6 hours. The mapping and re-gridding of the MOZART-4 data were conducted by matching the CMAQ grid information.

2.2 Observation data

2.2.1. Satellite-based observations

A Korean geostationary satellite of Communication, Ocean, and Meteorological Satellite (COMS) was launched on 26 June in 2010 over the Korean Peninsula. The COMS is a geostationary orbit satellite and it is stationed at an altitude of approximately 36,000 km at a latitude of 36°N and a longitude of 128.2°E with a horizontal coverage of 2500 × 2500 km^2 (refer to Fig. 1). Among the three payloads of the COMS, Geostationary Ocean Color Image

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(GOCI) is the first multi-channel ocean color sensor with visible and near infrared channels. The GOCI instrument provides hourly spectral images with a spatial resolution of 500 × 500 m^2 from 00:30 to 07:30 Coordinated Universal Time (UTC) for eight spectral (6 visible and 2 near-infrared) channels at 412, 443, 490, 555, 660, 680, 745, and 865 nm.

The Yonsei aerosol retrieval (YAER) algorithm for the GOCI sensor was initially developed by Lee et al. (2010) to retrieve the aerosol optical properties (AOPs) over ocean areas, and was then improved by expanding to consider non-spherical aerosol optical properties (Lee et al., 2012). Choi et al. (2016) further extended the algorithm for application to land surfaces, and the algorithm was referred to as the GOCI YAER version 1 algorithm. With the GOCI YAER algorithm, hourly Aerosol Optical Depths (AODs) at 550 nm were produced over East Asia. Choi et al. (2016) compared the retrieved GOCI AODs with other satellite-retrieved and ground-based observations, and found several errors in the cloud masking and surface reflectances. These errors were corrected in the recently updated second version of the GOCI YAER algorithm (Choi et al., 2018), which used the updated cloud masking and more accurate surface reflectances. In this study, the most recent GOCI AOD products from the GOCI YAER version 2 algorithm were used.

2.2.2. Ground-based observations

In addition to the satellite data, ground-based observations in South Korea and China were also collected for use in the operational air quality prediction system for PM and gas-phase pollutants. The orange, red, and blue dots in Fig. 1 represent the ground-based observation sites in China, Air Korea, and super-site stations in South Korea, respectively. These observations provide real-time concentrations of criteria species such as PM_{10}, $PM_{2.5}$,

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CO, O_3, SO_2, and NO_2.

Throughout the period of the KORUS-AQ campaign, ground-based observation data were obtained from 1514 stations in China, 264 Air Korea stations, and seven super-site stations in South Korea. In this study, 80 % of the ground-based observations in China and Air Korea stations in South Korea were randomly selected for use in the prediction system. The other 20 % of the data and super-site observations were used to evaluate the performances of the developed air quality prediction system.

In addition, AErosol RObotic NETwork (AERONET) AODs were used to conduct an independent evaluation of the air quality prediction system. AERONET is a federated global ground-based sun photometer network (Holben et al., 1998). Cloud-screened and quality-assured level 2.0 AODs for the AERONET were used in this study.

2.3 Operational air quality prediction system

In the present study, the operational air quality prediction system was developed by adjusting the ICs for the CMAQ model simulations based on DA with satellite-retrieved and ground-measured observations. Two parallel WRF-CMAQ model runs were conducted. The first experiment that involved adjusting ICs via DA is referred to as DA RUN (see Fig. 2). In order to evaluate the prediction system, a second experiment, in which the ICs were originated from the previous CMAQ model simulations without assimilations, was also conducted. This CMAQ run is referred to as BASE RUN.

2.3.1. AOD calculations

CMAQ AODs are calculated by integrating the aerosol extinction coefficient (σ_{ext})

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using the following equation:

$$\mathrm{AOD}(\lambda) = \int_0^z \sigma_{\mathrm{ext}}(\lambda)\ \mathrm{dz} \tag{1}$$

where z represents the vertical height; σ_{ext} is defined as the sum of the absorption coefficient (σ_{abs}) and the scattering coefficient (σ_{sca}); and σ_{abs} and σ_{sca} can be estimated by Eqns (3) and (4), respectively, as shown below:

$$\sigma_{\mathrm{ext}}(\lambda) = \sigma_{\mathrm{abs}}(\lambda) + \sigma_{\mathrm{sca}}(\lambda) \tag{2}$$

$$\sigma_{\mathrm{abs}}(\lambda)\ [\mathrm{Mm^{-1}}] = \sum_i^n \sum_j^m \{(1 - \omega_{ij}(\lambda)) \cdot \beta_{ij}(\lambda) \cdot f_{ij}(\mathrm{RH}) \cdot [\mathrm{C}]_{ij}\} \tag{3}$$

$$\sigma_{\mathrm{sca}}(\lambda)\ [\mathrm{Mm^{-1}}] = \sum_i^n \sum_j^m \{\omega_{ij}(\lambda) \cdot \beta_{ij}(\lambda) \cdot f_{ij}(\mathrm{RH}) \cdot [\mathrm{C}]_{ij}\} \tag{4}$$

where i and j denote the particulate species and size bin (or particle mode), respectively; $\omega_{ij}(\lambda)$ is the single scattering albedo; $\beta_{ij}(\lambda)$ is the mass extinction efficiency (MEE) of particulate species i for the size bin or particle mode j; $[\mathrm{C}]_{ij}$ is the concentration of particulate species including $(NH_4)_2SO_4$, NH_4NO_3, black carbon, organic aerosols (OA), mineral dust, and sea-salt aerosols; RH is the relative humidity; and $f_{ij}(\mathrm{RH})$ is the hygroscopic factor.

Here, the single scattering albedo (ω) refers to the fraction (portion) of the scattering over total extinction. In this work, σ_{ext} was estimated using β and $f(\mathrm{RH})$, as suggested by Chin et al. (2012). Park et al. (2014) and Lee et al. (2016) found that the values reported by Chin et al. (2012) produced the best results in estimating AODs at 550 nm over East Asia. The calculated AODs were used in the air quality prediction system in order to prepare the

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ICs for the PM predictions.

2.3.2. Data assimilation (DA)

The ground-based observations, together with GOCI-derived AODs, were used to prepare the ICs for the operational air quality predictions with the CMAQ model simulations. In order to achieve this, the following steps were taken: (i) the CMAQ-calculated concentrations of CO, O_3, and SO_2 were combined with the concentrations of CO, O_3, and SO_2 obtained from ground-based observations in South Korea (Air Korea) and China; (ii) the CMAQ-calculated AODs were assimilated with the GOCI AODs; (iii) the assimilated AODs were converted into PM_{10}; (iv) the converted PM_{10} was again assimilated at the surface in South Korea and China; and (v) after the DA at the surface, the ratios of the assimilated species concentrations to the original CMAQ-simulated concentrations were applied so as to the adjust vertical profiles of the chemical species above the surface. In the operational prediction system, the DA cycle is 24 hours and the assimilation takes place every day at 00:00 UTC (refer to Fig. 3).

The optimal interpolation (OI) method with the Kalman filter was chosen in the operational air quality prediction system. The OI method was originally used for meteorological applications (Lorenc, 1986), and has also been used in the assimilations for trace gases (Khattatov et al., 1999, 2000; Lamarque et al., 1999; Levelt et al., 1998). Recently, the OI technique has also been applied to aerosol fields (Collins et al., 2001; Yu et al., 2003; Generoso et al., 2007; Adhikary et al., 2008; Carmichael et al., 2009; Chung et al., 2010; Park et al., 2011; Tang et al., 2015, 2017).

Aerosol assimilation using the OI method was first applied by Collins et al. (2001) as

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follows:

$$\tau_m' = \tau_m + \mathbf{K}(\tau_o - \mathbf{H}\tau_m) \tag{5}$$

$$\mathbf{K} = \mathbf{B}\mathbf{H}^T(\mathbf{H}\mathbf{B}\mathbf{H}^T + \mathbf{O})^{-1} \tag{6}$$

$$\mathbf{O} = [(f_o\tau_o)^2 + (\varepsilon_o)^2]\mathbf{I} \tag{7}$$

$$\mathbf{B}(d_x, d_z) = [(f_m\tau_m)^2 + (\varepsilon_m)^2]\exp\left[-\frac{d_x^2}{2l_{mx}^2}\right]\exp\left[-\frac{d_z^2}{2l_{mz}^2}\right] \tag{8}$$

where τ_m', τ_m, and τ_o represent the assimilated products by the OI method, the modeled values, and the observed values, respectively; $\mathbf{K}$ is the Kalman gain matrix; $\mathbf{H}$ is the observation operator (or forward operator), which is an interpolator from model to observation space; $\mathbf{B}$ and $\mathbf{O}$ are the background and observation error covariance matrices, respectively; $(\cdot)^T$ denotes the transpose of a matrix; f_o is the fractional error in the observation-retrieved value; ε_o is the minimum root mean square error in the observation-retrieved values; f_m is the fractional error in the model estimates; ε_m is the minimum root mean square error in the model estimates; d_x is the horizontal distance between two model grid points; l_{mx} is the horizontal correlation length scale for the errors in the model; d_z is the vertical distance between two model grid points; and l_{mz} is the vertical correlation length scale for the errors in the model. In this work, the OI technique was applied for the DA of atmospheric gaseous species as well as particulate species.

Six free parameters (f_m, f_o, ε_m, ε_o, l_{mx}, and l_{mz}) were used to calculate the error covariance matrices of the observations and model, the mathematical formalisms of which are described in Eq. (7) and (8), respectively. Several previous studies have used fixed values for free parameters (Collins et al., 2001; Yu et al., 2003; Adhikary et al., 2008; Chung et al.,

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2010). These runs are called "static" runs. In contrast to those previous studies, "non-static" free parameters were applied in this study by minimizing the differences between the assimilated values and observations via an iterative process at each assimilation time step. This non-static free parameter scheme is possible due to the fact that the OI technique with the Kalman filter is much less costly in terms of computation time than other DA techniques, such as the 3-D or 4-D variational methods. This is another advantage of using the OI technique in this system. It typically takes less than 20 minutes with a workstation environment (dual Intel Xeon 2.40 GHz processor).

2.3.3. Allocation of the assimilated PM_{10} & $PM_{2.5}$ into particulate composition

In the procedure of operational DA, PM_{10} was assimilated in this study, because the PM_{10} data were more plentiful than $PM_{2.5}$. The assimilated PM_{10} then needs to be allocated into the PM composition for the CMAQ-model prediction runs. In order to achieve this, the differences between the assimilated PM_{10} and background PM_{10} (ΔPM_{10}) were first calculated. Then, $\Delta PM_{2.5}$ was estimated using the ratios of $PM_{2.5}$ to PM_{10} from the background CMAQ model runs (i.e., $\Delta PM_{2.5}=\Delta PM_{10}\times PM_{2.5}/PM_{10}$). $\Delta PM_{2.5}$ was then allocated to the $PM_{2.5}$ composition according to the comparison between two $PM_{2.5}$ compositions observed at the seven super-sites and simulated from the CMAQ model runs over South Korea. Both of the compositions are shown in Fig. 4. In Fig. 4, "PM OTHERS" indicates the remaining particulate matter species after excluding sulfate, nitrate, ammonium, organic aerosol (OA), and elementary carbon (EC). The PM OTHERS occupies 25 % of the total $PM_{2.5}$ observed at super-sites. The other fraction, $\Delta PM_{10}\times(1-PM_{2.5}/PM_{10})$, was also distributed into the coarse-mode particles ($PM_{2.5-10}$) as crustal elements.

3. Results and discussions

The performances of the air quality prediction system were evaluated by comparing them with ground-based observations from the Air Korea network and super-site stations in South Korea. Several sensitivity analyses were also conducted in order to assess the influences of the DA time-intervals on the accuracy of the air quality prediction.

3.1. Evaluation of the air quality prediction system

3.1.1. Time-series analysis

Figure 5 shows the time-series plots of PM_{10}, $PM_{2.5}$, CO, O_3, and SO_2 concentrations from the BASE RUN and the DA RUN. Here, the observation data (OBS) obtained from the Air Korea network were compared with the results of the two sets of the CMAQ model simulations, i.e., (1) BASE RUN and (2) DA RUN. As mentioned previously, 20% of the Air Korea observations used in the evaluation were randomly selected during the period of the KORUS-AQ campaign. The other 80 % of the Air Korea data were used in the DA at 00:00 UTC. For the forecast hours from 01:00 to 23:00 UTC, all of the ground observations (254 Air Korea and seven super-site stations) were used to evaluate the performances of the developed air quality prediction system. As shown in Fig. 5, we achieved some improvements in the prediction performances by applying the ICs to the CMAQ model simulations. The BASE RUN significantly under-predicted PM_{10}, $PM_{2.5}$, and CO while the DA RUN produced concentrations that were more consistent with the observations than those of the BASE RUN.

In case of CO, the observed CO mixing ratios were about three times higher than those from the BASE RUN. These large differences are well known, and have been attributed to the underestimated emissions of CO (Heald et al., 2004). However, when the DA was

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applied, the predictions of the CO mixing ratios improved. Similarly, the performances of the PM_{10} and $PM_{2.5}$ predictions improved with the application of the DA. Unlike PM_{10}, $PM_{2.5}$, and CO, the O_3 mixing ratios and its diurnal trends from both the BASE RUN and DA RUN tend to be well-matched with the observations. By contrast, the poorest performances of the BASE RUN and the DA RUN were shown for SO_2.

In addition, a dust event took place between 6 May and 8 May. This event is captured by the DA RUN (check red peaks in Fig. 5(a) and (b)), while the BASE RUN cannot capture this dust event. This demonstrates the capability of the current system to possibly predict dust events in South Korea. In the DA RUN, dust information is provided into the CMAQ model runs through both/either GOCI AOD and/or ground PM observations measured along the dust plume tracks.

The effectiveness of the DA with prediction time was also analyzed by calculating the aggregated average concentrations of atmospheric species (see Figs. 6, 7, and 9). Fig. 6 depicts the CMAQ-calculated average concentrations of PM_{10}, $PM_{2.5}$, CO, and SO_2 against the Air Korea observations. Our air quality prediction system re-generated relatively well-matched concentrations for PM_{10}, $PM_{2.5}$, and CO from the DA RUN.

Figure 7 shows the case of ozone. The ozone mixing ratios from the DA RUN in Fig. 7(a) were reasonably consistent with the observations at 00:00 UTC, but disagreed with those between 04:00 and 09:00 UTC (13:00 and 18:00 KST), when solar insolation is the most intense. This may be attributed to the chemical imbalances between ozone production and ozone destruction (or titration). However, if CMAQ NO_2 was assimilated with ground-based observations in South Korea (Air Korea) and China, the predicted ozone mixing ratios became substantially closer to the observations, as shown in Fig. 7(b). This is clearly due to

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the fact that NO_x is an important precursor of ozone. In the prediction of the ozone mixing ratios, both 1-hr peak ozone (around 15:00 KST) and 8-hr averaged ozone mixing ratios (between 9:00 and 17:00 KST) are important. Fig. 7 clearly shows that the prediction accuracies of both the ozone mixing ratios were improved after the DA of NO_2 mixing ratios.

Although the DA for NO_2 provided better ozone predictions, one should take caution in using the NO_2 observations. The NO_2 mixing ratios measured at Air Korea sites are known to be contaminated by other nitrogen gases such as nitric acid (HNO_3), peroxyacetyl nitrates (PANs), and alkyl nitrates (ANs), since the Air Korea NO_2 mixing ratios are measured through a chemiluminescent method with catalysts of gold or molybdenum oxide at high temperatures. These are known to be "NO_2 measurement artifacts" (Jung et al., 2017), which is one of the reasons that the DA of NO_2 was not shown in Fig. 6. The NO_2 mixing ratios are corrected from the Air Korea NO_2 data, and are then used to prepare the ICs via the DA for more accurate ozone and NO_2 predictions. Currently, such corrections of the observed NO_2 mixing ratios are being standardized with more sophisticated year-long NO_2 measurements. After the corrections of the NO_2 measurement artifacts, more evolved schemes of ozone and NO_2 predictions will be possible in the future. As shown in Fig. 7, about a 20% reduction (average fraction of non-NO_2 mixing ratios in the observed NO_2 mixing ratios) was made for these demonstration runs (Jung et al., 2017).

Another practical issue is now discussed. Although the assimilation with the observed NO_2 mixing ratios can enhance the accuracy of the predictions of the daytime ozone mixing ratios, the nighttime ozone mixing ratios tend to be consistently over-predicted in the aggregated plot of the ozone mixing ratios at the observation sites (see Fig. 7). This is believed to be caused by underestimation of the mixing layer height (MLH). Figure 8 shows a

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comparison between lidar-measured MLH (black dashed line) and WRF-calculated MLH (with the option of the Yonsei University (YSU) planetary boundary layer scheme) (Hong et al., 2006; see red line). As shown in Fig. 8, the nocturnal lidar-measured MLH is about two times higher than the nocturnal WRF-calculated MLH as measured at a lidar site inside the campus of Seoul National University (SNU) in Seoul. This is a common and well-defined phenomenon in East Asia. Such underestimated MLH in the model tends to compress the ozone molecules within the mixing layer during the nighttime, which leads to consistently over-predicted nocturnal ozone mixing ratios.

Although the correct predictions of the daytime ozone mixing ratios are substantially more important, it is also worth trying to achieve correct predictions of the nocturnal ozone mixing ratios. Correct predictions of the nocturnal ozone mixing ratios strongly depend on the correct estimation of the MLH. Currently, efforts are being made in two directions. First, a modified MLH (or PBL) scheme in the meteorological model is currently being studied. The other area of study is that the WRF-calculated MLH can be "bias-corrected" to match the observed MLHs in the interface (MCIP) between a MET model (e.g., WRF) and a CTM model (e.g., CMAQ). These efforts are now underway as well.

In this work, the aerosol composition (such as EC, OA, sulfate, nitrate, and ammonium) was further compared with the composition observed at the super-sites shown in Fig. 9. As shown in Fig. 9, agreement was observed between the DA RUN and observations for all of the major PM constituents. Again, this is another strong capability of our system for predicting not only particle mass, but also the "chemical composition" of particulate matters.

3.1.2. Spatial distribution

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Figure 10 shows the spatial distributions of PM and chemical species throughout the entire period of the KORUS-AQ campaign over the Seoul Metropolitan Area (SMA). Noticeable improvements are observed to have been achieved in the spatial distributions by applying the ICs into the CMAQ model simulations, particularly for PM_{10} (Fig. 10a), $PM_{2.5}$ (Fig. 10b), and CO (Fig. 10c). As shown in Fig. 10, the under-predicted concentrations of PM_{10}, $PM_{2.5}$, and CO were adjusted to concentrations closer to the observations. In case of SO_2 (see Fig. 10d), the DA RUN produced better agreement with the observations than the BASE RUN, but there were still under-predicted SO_2 concentrations over the northeastern part of the SMA.

By contrast, relatively lower ozone mixing ratios from the DA RUN against the BASE RUN were found in the southwestern part of the SMA (see Fig. 10e). Due to the nonlinear relationship between NO_x and O_3, high mixing ratios (or emissions) of NO_x in the SMA can lead to depletion of ozone. In these runs, the precursors of ozone such as NO_x and VOCs were excluded in the preparation of the ICs for CMAQ model simulations. Again, this is because the Air Korea NO_2 mixing ratios are contaminated by several reactive nitrogen species, so the data cannot be directly used in the assimilation procedures. In case of VOCs, a limited number of datasets is available in South Korea for the DA. Improvements in the prediction of ozone mixing ratios can be achieved when the NO_2 mixing ratios are corrected and a sufficient number of VOCs data (possibly from satellite data in the future) is available.

3.1.3. Statistical analysis

In order to achieve better understanding of the performances of the DA RUN, analyses of statistical variables such as index of agreement (IOA), Pearson's correlation

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coefficient (R), root mean square error (RMSE), and mean bias (MB) were conducted using observations from the Air Korea stations for PM_{10}, $PM_{2.5}$, CO, SO_2, and O_3 (see Fig. 11). Definitions of the statistical variables are given in Appendix A.

After the applications of the ICs, both RMSE and MB became lower, while the correlation coefficient became higher for the entire predictions. In addition, it was found that the differences between the BASE RUN and the DA RUN tended to diminish as the prediction time progressed. The results of the statistical analysis are listed in Table 1. The results of the DA RUN were reasonably consistent with the observations for PM_{10} (IOA = 0.60; R= 0.40; RMSE = 34.87; MB = -13.54) and $PM_{2.5}$ (IOA = 0.71; R= 0.53; RMSE = 17. 83; MB = -2.43), as compared to the BASE RUN for PM_{10} (IOA = 0.51; R= 0.34; RMSE = 40.84; MB = -27.18) and $PM_{2.5}$ (IOA = 0.67; R= 0.51; RMSE = 19.24; MB = -9.9). In terms of bias, an improvement was found for CO: MB = -0.036 for the DA RUN and MB = -0.27 for the BASE RUN. Regarding O_3 and SO_2, the DA RUN showed slightly better performances than the BASE RUN.

Table 2 presents the results of the statistical analysis at 00:00 UTC when the DA was conducted, with the results clearly showing how much closer the DA makes the CMAQ-calculated chemical concentrations to the observed concentrations. Collectively, the DA improved model accuracy by a large degree in terms of R, particularly for PM_{10} (R: 0.3→0.75; slope: 0.17→0.66) and O_3 (R: 0.09→0.61; slope: 0.07→0.42). In addition, for all species, MB and RMSE decreased significantly with the DA RUN as compared with the BASE RUN.

3.2. Sensitivity test of DA time-interval

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3.2.1. AOD

In this section, a sensitivity analysis was conducted with different implementation time-intervals of the DA (i.e., 24, 6, and 3 hours) for AOD (refer to Fig. 12). As shown in Fig. 12, more frequent implementation of the DA is expected to make the predicted results closer to the observations. Although the DA RUN with a shorter assimilation time-interval tends to produce a better prediction, it is not always the most appropriate choice, since the shorter assimilation time-interval results in increased computational cost. Therefore, an optimized assimilation time-interval should be found to achieve the best performances from the given DA system with the consideration of its own computational ability.

3.2.2. PM and gases

In addition, sensitivity analyses of the developed air quality prediction system to multiple implementations of the DA with different time-intervals were also investigated for (a) PM_{10}, (b) $PM_{2.5}$, (c) CO, (d) SO_2, and (e) O_3, shown in Fig. 13. Fig. 13 shows a soccer plot analysis for BASE RUN (blue crosses) and DA RUNs with different DA time-intervals of 24 hours (OI; red circles), two hours (2-hr OI; black diamonds), and one hour (1-hr OI; dark-green triangles). This set of testing was designed based on the fact that the performances are expected to improve if the DAs are implemented multiple times prior to the actual predictions at 00:00 UTC. Here, for the 2-hr OI run, the DA was implemented three times a day at 20:00, 22:00, and 00:00 UTC, while for the 1-hr OI run, the DA was implemented at 22:00, 23:00, and 00:00 UTC. The performances of all of the chemical species excluding ozone improved, as expected, with DA RUNs with more frequent and longer DA time-intervals (i.e., three-times implementation with a 2-hr time-interval in our cases). In case of ozone, the best

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performance was found for the air quality prediction system with the DA time-interval of 24-hr.

Unsurprisingly, more frequent DAs prior to the actual prediction mode (i.e., before 00:00 UTC in our system) with a longer time-interval (such as 2-hr) will be computationally costly. There will certainly be a "trade-off" between the precision of air quality prediction and the computational cost. The system should be designed under the consideration of these two factors.

4. Summary and conclusions

In this study, an operational air quality prediction system was developed by preparing the ICs for CMAQ model simulations using GOCI AODs and ground-based observations of PM_{10}, CO, ozone, and SO_2 during the period of the KORUS-AQ campaign (1 May – 12 June 2016) in South Korea. The major advantages of the developed air quality prediction system are its comprehensiveness in predicting the ambient concentrations of both gaseous and particulate species (including PM composition) as well as its powerfulness in terms of computational cost.

The performances of the developed prediction system were evaluated using ground in-situ observation data. The CMAQ model runs with the ICs (DA RUN) showed higher consistency with the observations of almost all of the chemical species, including PM composition (sulfate, nitrate, ammonium, OA, and EC) and atmospheric gases (CO, ozone, and SO_2), than the CMAQ model runs without the ICs (BASE RUN). Particularly for CO, the DA was able to remarkably improve the model performances, while the BASE RUN significantly under-predicted the CO concentrations (predicting about one-third of the

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observed values). In case of ozone, both the BASE RUN and DA RUN were in close agreement with observations. More reliable predictions of ozone mixing ratios will be achieved via the DA of the observed NO_2 mixing ratios and the corrections of model-simulated mixing layer height (MLH). For SO_2, the performances of both the BASE RUN and the DA RUN were somewhat poor. Regarding this issue, more accurate SO_2 emissions are required to achieve better SO_2 predictions, and these can be estimated through inverse modeling using satellite data (e.g., Lee et al., 2011). The adjustments of both ICs and emissions may be able to improve the performances of the air quality prediction system, and this will be examined in future studies.

Moreover, the developed air quality prediction system will be upgraded by using the new observation data that will be retrieved after 2020 from the Geostationary Environment Monitoring Spectrometer (GEMS) with a high spatial resolution of 7×8 km^2 as well as a high temporal resolution of 1-hour over a large part of Asia. In addition, the current DA technique of the OI with the Kalman filter can also be upgraded with the use of more advanced DA methods such as variational techniques of 3DVAR and 4DVAR methods, as well as with the ensemble Kalman filter (EnK) method. These research endeavors are currently underway.

In conjunction with improving the air quality modeling system, artificial intelligence (AI)-based air quality prediction systems are also currently being developed in several ways (e.g., H. S. Kim et al., 2019). Both the CTM-based and AI-based air quality prediction systems will be combined so as to ultimately enable more accurate air quality forecasts over South Korea for Korean citizens. This is the ultimate goal of our research.

Code and data availability. WRF v3.8.1 (doi:10.5065/D6MK6B4K) and CMAQ v5.1 (doi:10.5281/zenodo.1079909) models are both open-source and publicly available. Source codes for WRF and CMAQ can be downloaded at http://www2.mmm.ucar.edu/wrf/users/downloads.html and https://github.com/USEPA/CMAQ, respectively. Data from the KORUS-AQ field campaign can be downloaded from the KORUS-AQ data archive (http://www-air.larc.nasa.gov/missions/korus-aq). Other data were acquired as follows. Ground-based observation data were downloaded from the Air Korea website (http://www.airkorea.or.kr) for South Korea and https://pm25.in for China. AERONET data were downloaded from https://aeronet.gsfc.nasa.gov. All codes related with the air quality prediction system can be obtained by contacting K. Lee (lkh1515@gmail.com).

Author contributions. KL developed the model code, performed the simulations, and analyzed the results. CHS directed the experiments. JY contributed to shape the research and analysis. SL, MP, HH, and SYP helped analyze the results. MC, JK, YK, JHW, and SWK provided and analyzed data applied in the experiments. KL prepared the manuscript with contributions from all co-authors.

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APPENDIX A: FORMULAS FOR STATISTICAL EVALUATION INDICES

The formulas used to evaluate the performances of the operational air quality prediction system are defined as follows.

$$\text{Index Of Agreement (IOA)} = 1 - \frac{\sum_1^n (\mathrm{M}-\mathrm{O})^2}{\sum_1^n (|\mathrm{M}-\bar{\mathrm{O}}| + |\mathrm{O}-\bar{\mathrm{O}}|)^2} \quad \text{(A1)}$$

$$\text{Correlation Coefficient (R)} = \frac{1}{(\mathrm{n}-1)} \sum_1^n \left(\left(\frac{\mathrm{O}-\bar{\mathrm{O}}}{\sigma_\mathrm{O}} \right) \left(\frac{\mathrm{M}-\bar{\mathrm{M}}}{\sigma_\mathrm{m}} \right) \right) \quad \text{(A2)}$$

$$\text{Root Mean Square Error (RMSE)} = \sqrt{\frac{\sum_1^n (\mathrm{M}-\mathrm{O})^2}{\mathrm{n}}} \quad \text{(A3)}$$

$$\text{Mean Bias (MB)} = \frac{1}{\mathrm{n}} \sum_1^n (\mathrm{M}-\mathrm{O}) \quad \text{(A4)}$$

$$\text{Mean Normalized Bias (MNB)} = \frac{1}{\mathrm{n}} \sum_1^n \left(\frac{\mathrm{M}-\mathrm{O}}{\mathrm{O}} \right) \times 100\ \% \quad \text{(A5)}$$

$$\text{Mean Normalized Error (MNE)} = \frac{1}{\mathrm{n}} \sum_1^n \left(\frac{|\mathrm{M}-\mathrm{O}|}{\mathrm{O}} \right) \times 100\ \% \quad \text{(A6)}$$

$$\text{Mean Fractional Bias (MFB)} = \frac{1}{\mathrm{n}} \sum_1^n \frac{(\mathrm{M}-\mathrm{O})}{\left(\frac{\mathrm{M}+\mathrm{O}}{2} \right)} \times 100\ \% \quad \text{(A7)}$$

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$$\text{Mean Fractional Error (MFE)} = \frac{1}{n}\sum_{1}^{n}\frac{|M-O|}{\left(\frac{M+O}{2}\right)} \times 100\ \% \quad \text{(B8)}$$

In Eqns. (A1) - (A8), M and O represent the model and observation data, respectively. N is the number of data points and σ means the standard deviation. The overbars in the equations indicate the arithmetic mean of the data. The units of RMSE and MB are the same as the unit of data, while IOA and R are dimensionless statistical parameters.

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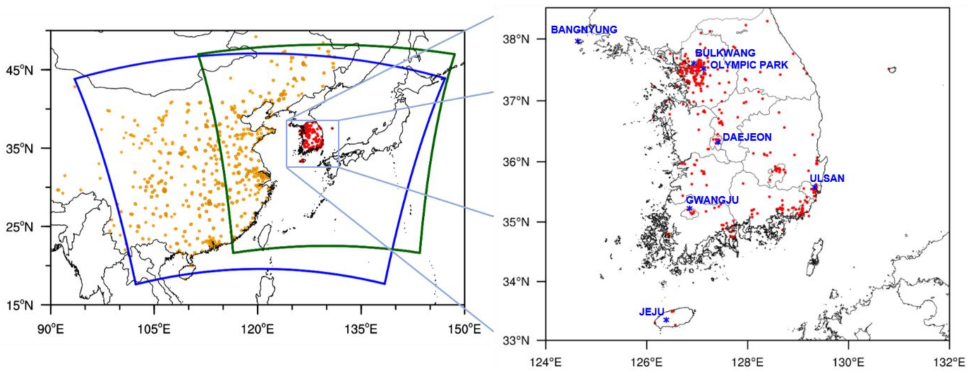

Figure 1. Domains of GOCI sensor (dark green line) and CMAQ model simulations (blue line). Red-colored dots denote the locations of Air Korea sites in South Korea. Orange-colored dots represent the locations of ground-based observation stations in China. Blue stars show the locations of seven super-sites in South Korea. During the KORUS-AQ campaign, observation data were obtained from 1514 stations in China as well as 264 Air Korea and seven super-site stations in South Korea.

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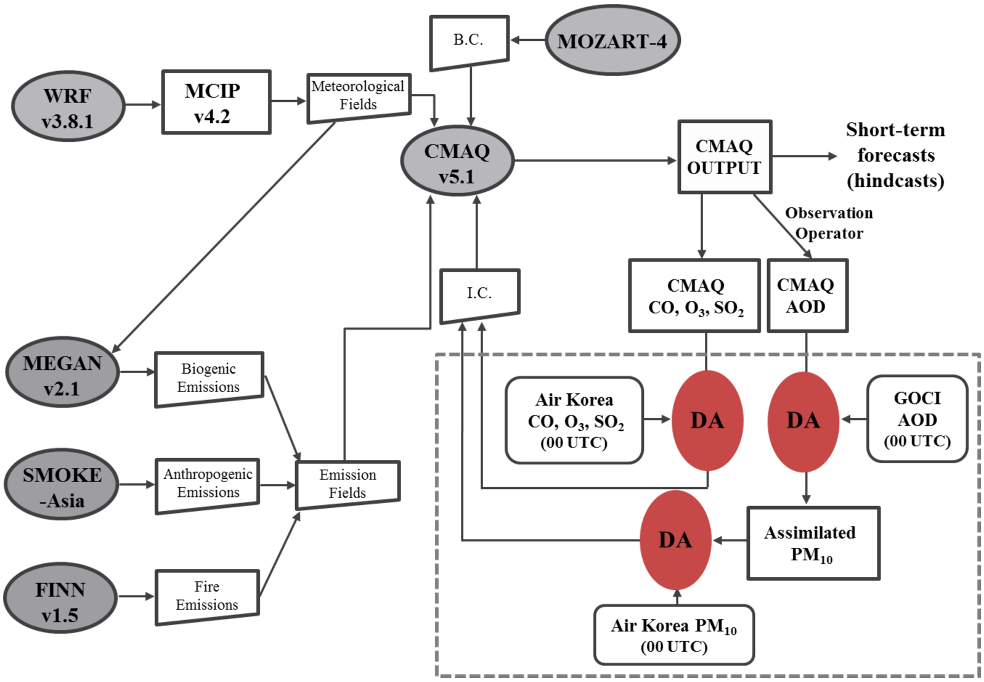

Figure 2. Schematic diagram of the Korean air quality prediction system developed in this study. The initial conditions (ICs) of the CMAQ model simulations are prepared by assimilating CMAQ outputs with satellite-retrieved and ground-measured observations. The data process for preparing the ICs is shown in the box with gray-dashed lines.

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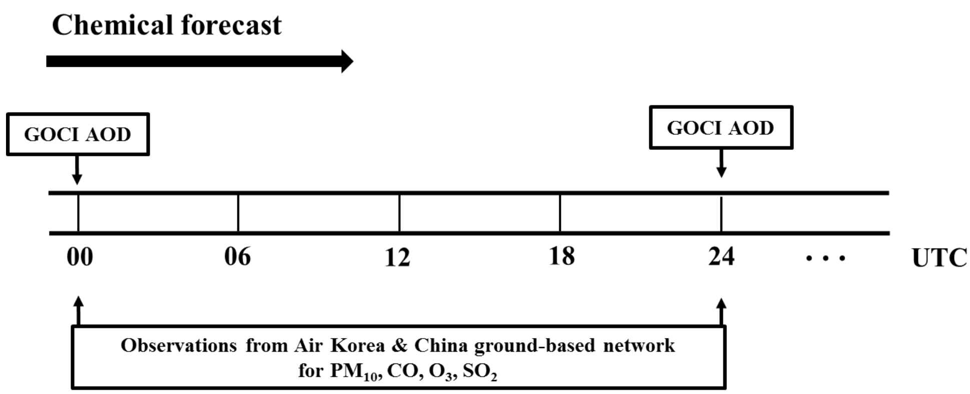

Figure 3. Schematic diagram of the Korean air quality prediction system for particulate matter (PM) and gas-phase pollutants. The data assimilation (DA) cycle is 24 hours for both PM and gas-phase pollutants such as CO, O_3, and SO_2. The DA of NO_2 is excluded in the current study, the reason for which is discussed in the text.

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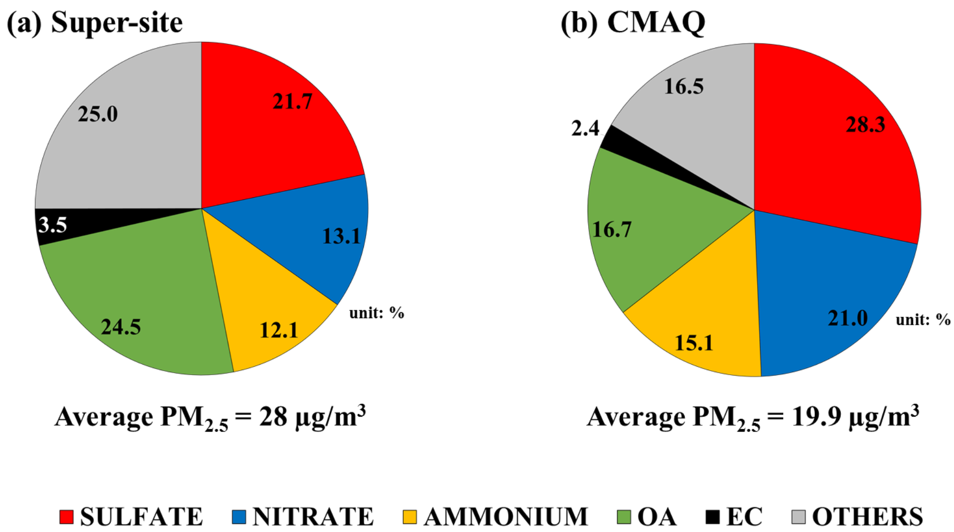

Figure 4. Average $PM_{2.5}$ composition (a) observed at the super-site stations and (b) simulated by the CMAQ model during the KORUS-AQ campaign. The averaged $PM_{2.5}$ measured from the super-sites and calculated from the CMAQ model simulations over the period of the KORUS-AQ campaign are 28 μg/m^3 and 19.9 μg/m^3, respectively. The mass of organic aerosols (OAs) was calculated by multiplying organic carbon mass by 1.6.

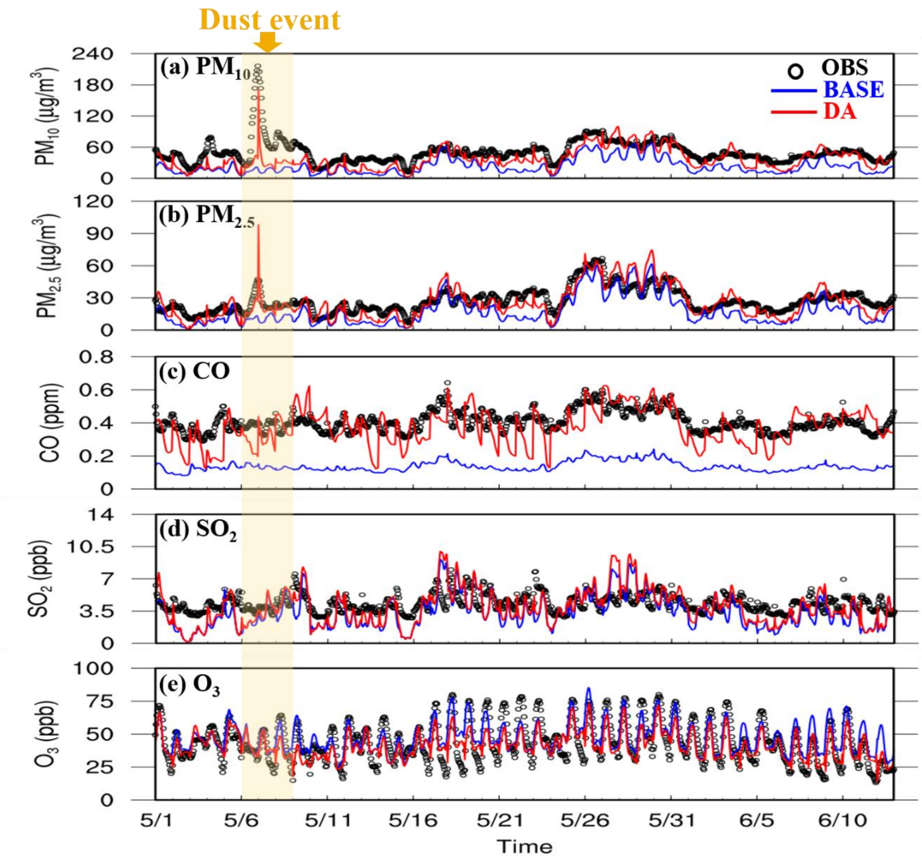

Figure 5. Time-series plots of hourly (a) PM_{10}, (b) $PM_{2.5}$, (c) CO, (d) SO_2, and (e) O_3 concentrations at 264 Air Korea stations. Black open circles (OBS) represent the observed concentrations. Blue and red lines show the results simulated from the BASE RUN and DA RUN over South Korea, respectively.

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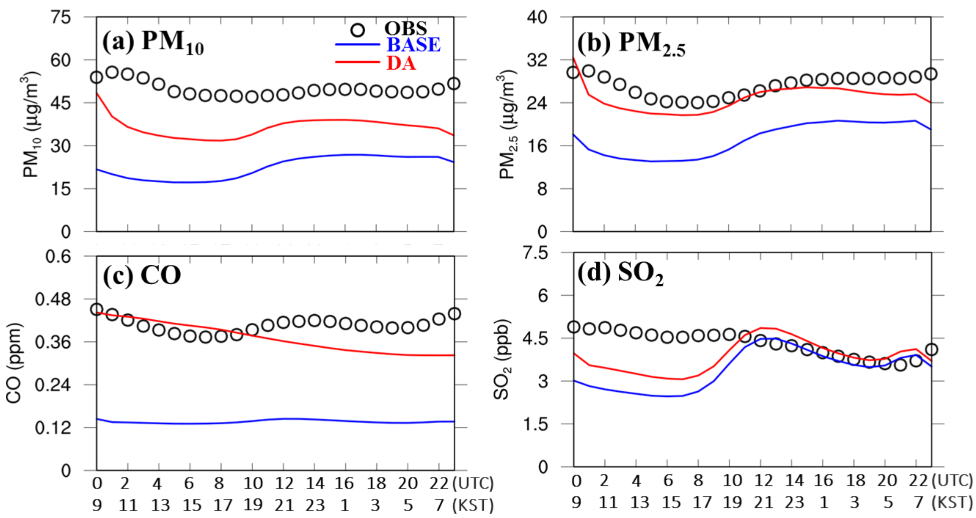

Figure 6. Aggregated average concentrations of (a) PM_{10}, (b) $PM_{2.5}$, (c) CO, and (d) SO_2 at 264 Air Korea stations over the KORUS-AQ campaign period. Open black circles denote the observations obtained from 264 Air Korea stations in South Korea. Blue and red lines represent the predicted concentrations from the BASE RUN and DA RUN, respectively. The DA was conducted at 00:00 UTC every day throughout the KORUS-AQ campaign period.

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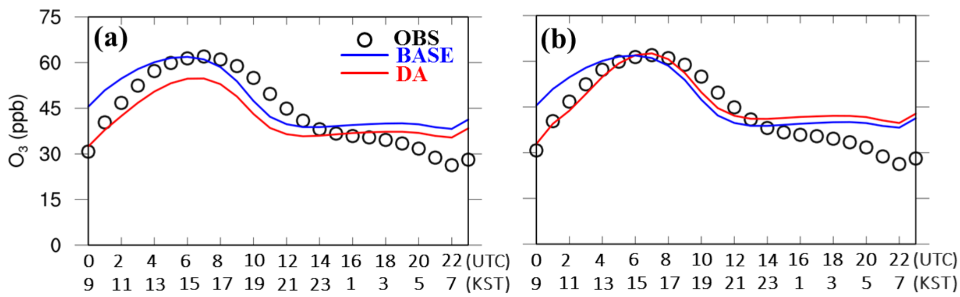

Figure 7. Comparison of CMAQ-simulated O_3 mixing ratios (BASE RUN with blue lines and DA RUN with red lines) with O_3 mixing ratios from Air Korea stations (open black circles). DA RUN was carried out by assimilating CMAQ outputs with Air Korea observations using (a) only O_3 mixing ratios and (b) both O_3 and NO_2 mixing ratios.

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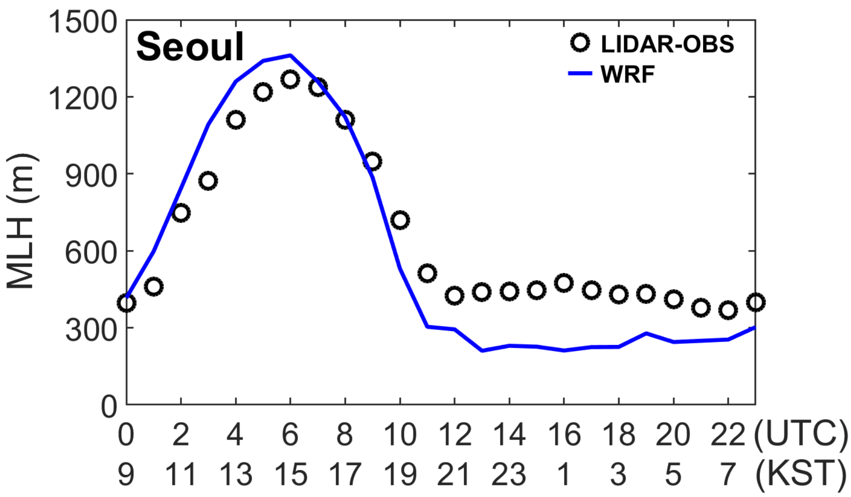

Figure 8. Comparison of WRF-simulated mixing layer height (MLH) (denoted by blue-dashed line) with lidar-measured MLH (denoted by open black circles) at Seoul National University (SNU) in Seoul. KST stands for Korean standard time.

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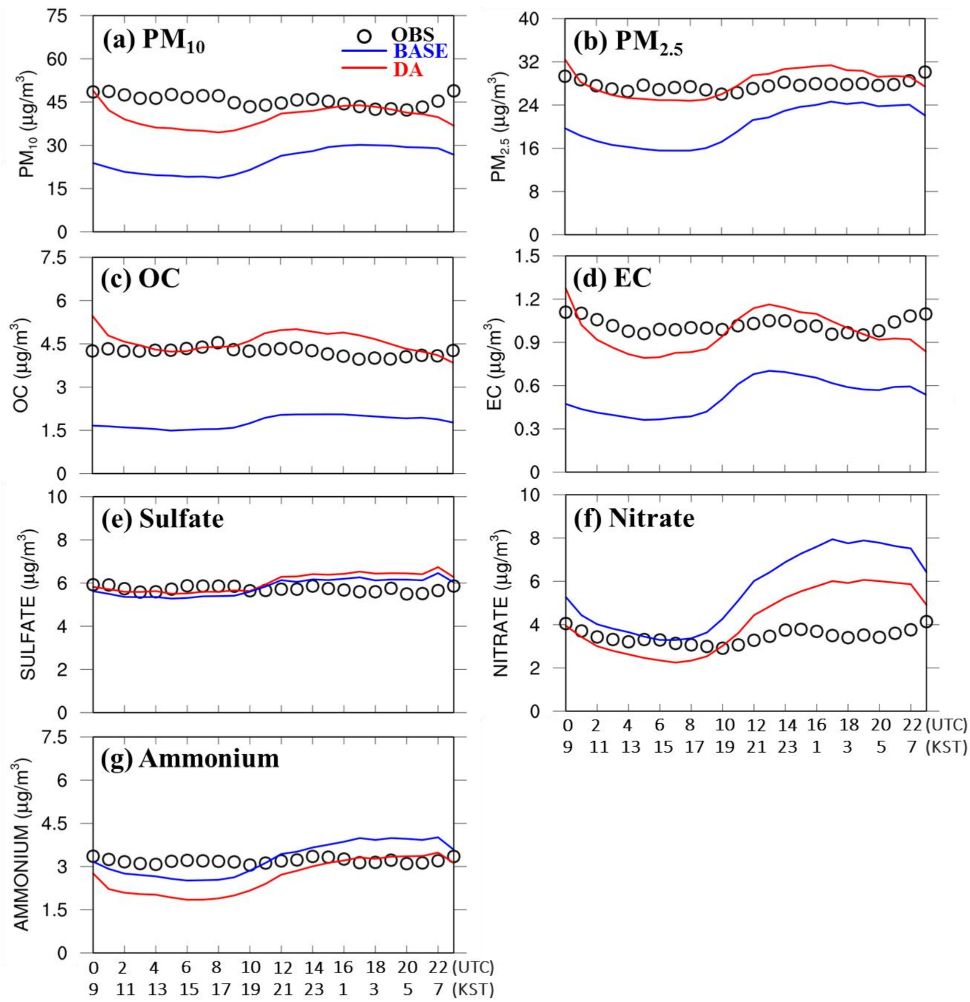

Figure 9. Aggregated average concentrations of (a) PM_{10}, (b) $PM_{2.5}$, (c) OC, (d) EC, (e) sulfate, (f) nitrate, and (g) ammonium as predicted by CMAQ model during the period of the KORUS-AQ campaign. The others are the same as those shown in Fig. 7, except for the fact that the observation data used here were obtained from the seven super-site stations in South Korea.

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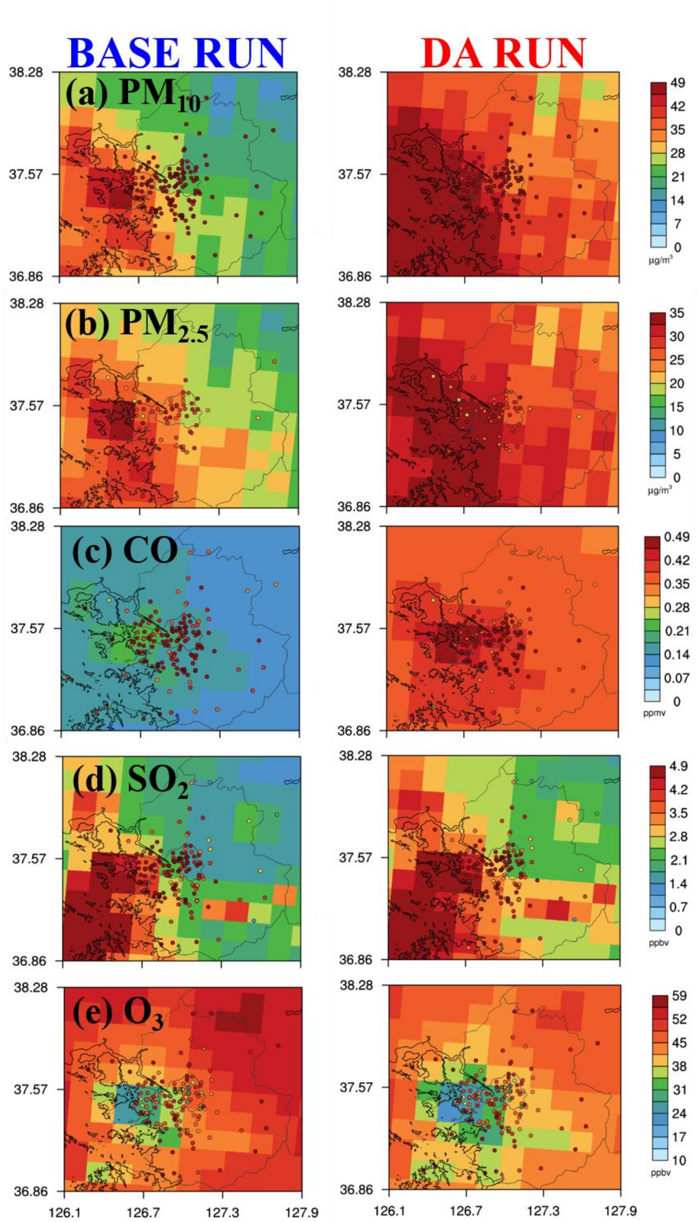

Figure 10. Spatial distributions of (a) PM_{10}, (b) $PM_{2.5}$, (c) CO, (d) SO_2, and (e) O_3 over Seoul Metropolitan Area (SMA). The concentrations were averaged over the entire period of the KORUS-AQ campaign. Colored circles represent the concentrations of the air pollutants observed at the Air Korea stations in the SMA.

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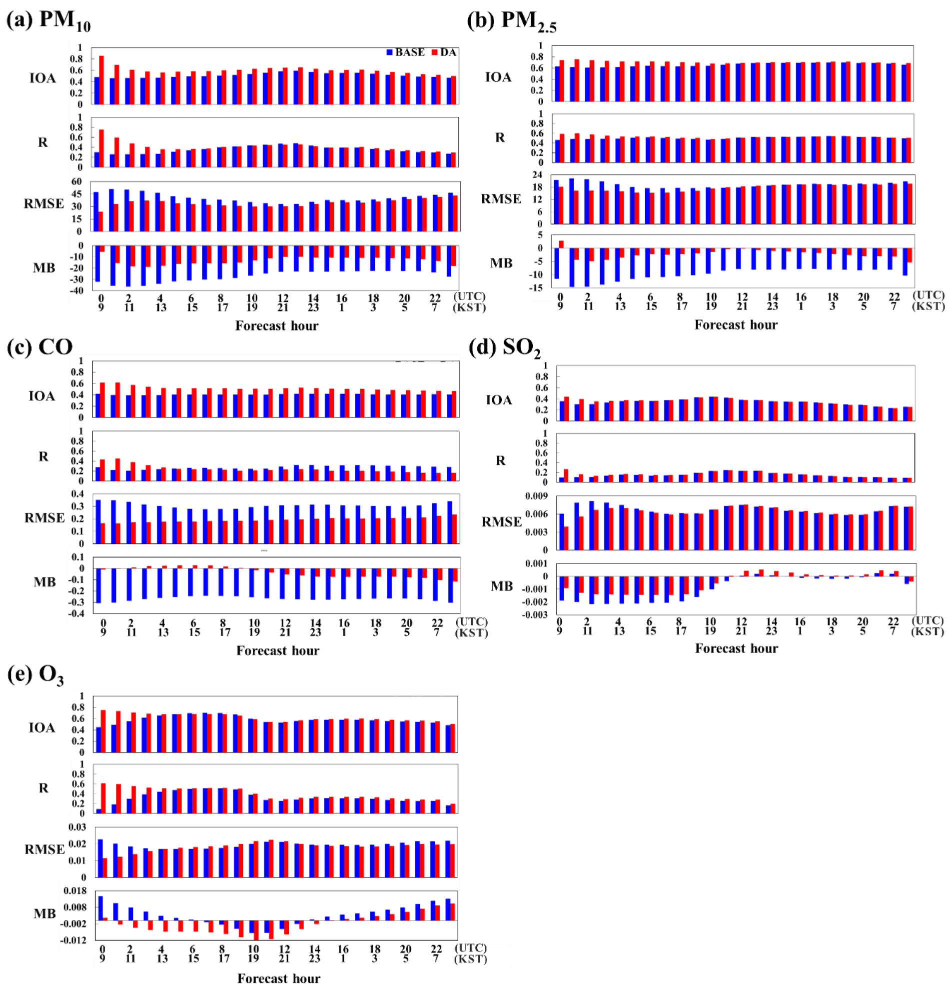

Figure 11. Time-series plots of four performance metrics (IOA, R, RMSE, and MB) for (a) PM_{10}, (b) $PM_{2.5}$, (c) CO, (d) SO_2, and (e) O_3 forecasts. The DA was conducted at 00:00 UTC. The units of RMSE and MB are $\mu g/m^3$ and ppmv for PM concentrations and for gaseous species, respectively. The definitions of the four performance metrics are shown in Appendix A.

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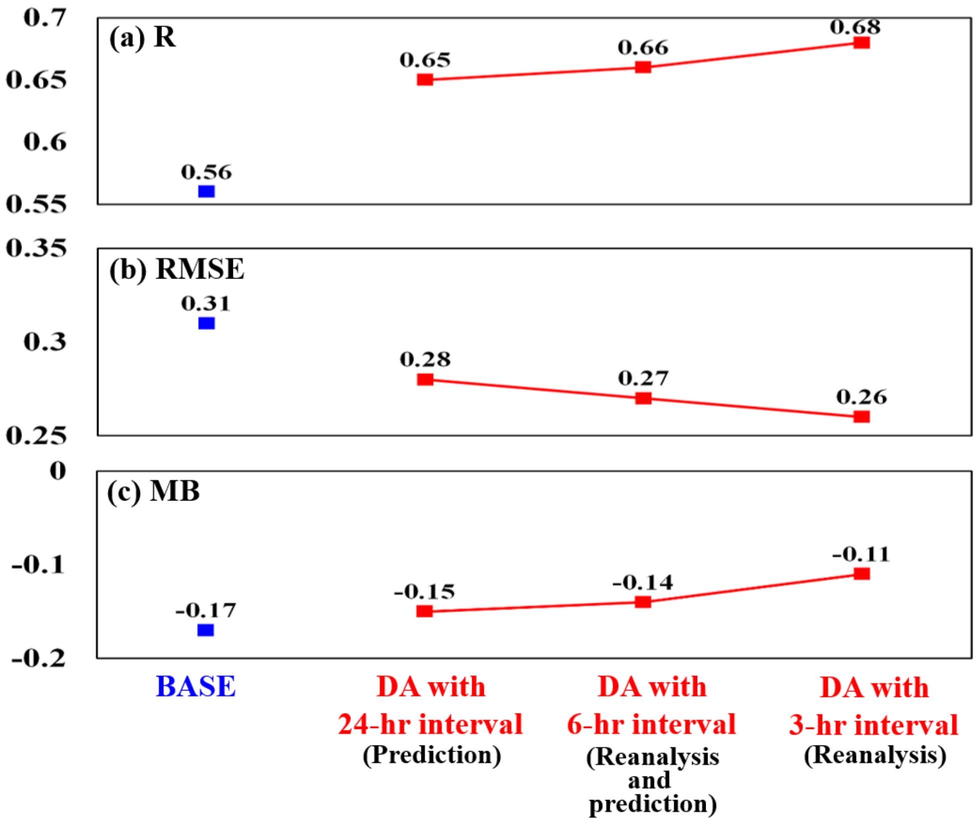

Figure 12. Variations of three performance metrics (R, RMSE, and MB) with time-intervals of data assimilations. For these tests, the GOCI AODs were used in the DA to update the initial conditions of the CMAQ model simulations. The results from the three CMAQ model simulations were compared with AERONET AODs ("ground truth"). The two blue squares represent the performances from the BASE RUNs and the red squares indicate the performances from the DA RUNs. The three experiments were carried out with the assimilation time-intervals of 24, 6, and 3 hours (hr), respectively. Here, the DA RUN with the 24-hr time-interval is referred to as "air quality prediction", and the DA RUNs with the 6-hr and 3-hr time-interval are referred to as "air quality reanalysis".

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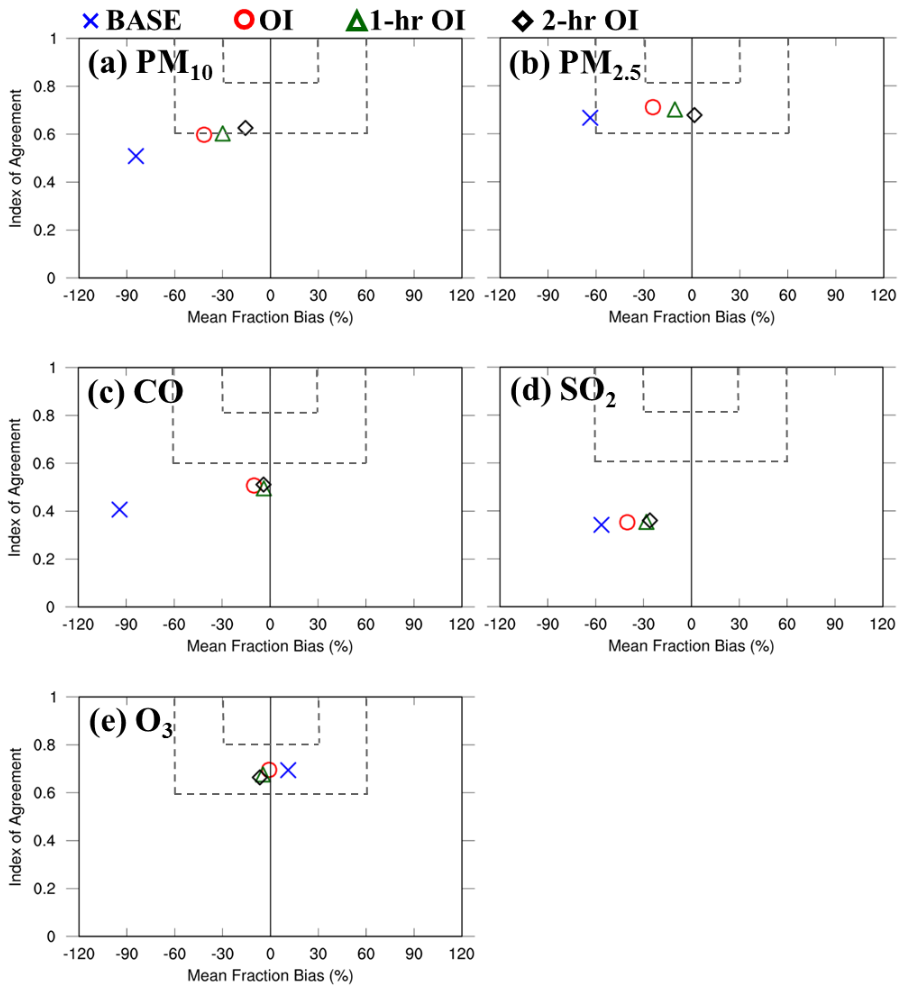

Figure 13. Soccer plot analyses for (a) PM_{10}, (b) $PM_{2.5}$, (c) CO, (d) SO_2, and (e) O_3. The CMAQ-predicted concentrations were compared with the Air Korea observations. Blue crosses, red circles, dark-green triangles, and black diamonds represent the performances calculated from the BASE RUN, the DA RUNs with the OI system, the 1-hour (hr) OI system, and the 2-hr OI system, respectively.

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Table 1. Statistical metrics from BASE RUN and DA RUN with Air Korea observations over the entire period of the KORUS-AQ campaign.

	PM_{10}		$PM_{2.5}$		CO		SO_2		O_3	
	BASE RUN	**DA RUN**	**BASE RUN**	**DA RUN**	**BASE RUN**	**DA RUN**	**BASE RUN**	**DA RUN**	**BASE RUN**	**DA RUN**
N	101852		65383		101764		101764		101836	
IOA	0.51	0.60	0.67	0.71	0.41	0.51	0.34	0.35	0.69	0.70
R	0.34	0.40	0.51	0.53	0.28	0.21	0.14	0.15	0.50	0.52
RMSE	40.8	34.87	19.2	17.83	0.31	0.19	0.0068	0.0066	0.020	0.02
MB	-27.2	-13.54	-9.9	-2.43	-0.27	-0.04	-0.0009	-0.0004	0.003	-0.0024
ME	30.1	24.20	15.3	13.48	0.27	0.15	0.004	0.0034	0.015	0.015
MNB	-50.0	-18.17	-30.1	5.32	-62.0	3.14	3.1	17.77	48.0	30.22
MNE	60.7	52.35	62.6	62.77	62.9	40.67	93.1	93.56	70.2	61.34
MFB	-84.3	-41.61	-63.6	-24.41	-94.1	-10.00	-56.4	-40.20	11.1	-0.82
MFE	91.1	62.32	81.6	60.01	94.9	39.49	91.4	82.91	40.7	40.64

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839 **Table 2.** Statistical metrics from BASE RUN and DA RUN with Air Korea observations at
840 00:00 UTC when the DA was conducted during the KORUS-AQ campaign.

	PM_{10}		$PM_{2.5}$		CO		SO_2		O_3	
	BASE RUN	**DA RUN**	**BASE RUN**	**DA RUN**	**BASE RUN**	**DA RUN**	**BASE RUN**	**DA RUN**	**BASE RUN**	**DA RUN**
N	1057		695		1024		1007		1043	
IOA	0.48	0.86	0.63	0.74	0.41	0.62	0.36	0.44	0.45	0.75
R	0.30	0.75	0.46	0.59	0.28	0.43	0.097	0.27	0.09	0.61
RMSE	47.2	23.92	21.5	18.21	0.35	0.16	0.0061	0.0039	0.023	0.012
MB	-32.2	-5.46	-11.5	2.80	-0.31	-0.01	-0.0019	-0.0009	0.015	0.002
ME	34.5	16.03	17.2	13.25	0.31	0.12	0.0039	0.0023	0.018	0.009
MNB	-54.9	-0.53	-33.2	26.17	-64.3	9.69	-20.1	7.35	100.4	27.45
MNE	64.0	36.07	63.1	59.77	64.8	30.69	86.7	55.27	107.8	43.81
MFB	-92.8	-13.38	-67.3	0.56	-98.7	1.81	-75.9	-17.39	43.7	12.16
MFE	98.8	38.41	84.3	48.30	99.1	27.14	99.9	56.23	52.9	31.53

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