Development of Korean Air Quality Prediction System version 1 (KAQPS v1) with focuses on practical issues

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24	Short title: Air quality prediction system in Korea
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28 Abstract

For the purpose of providing reliable and robust air quality predictions, an air quality 29 30 prediction system was developed for the main air quality criteria species in South Korea (PM₁₀, PM_{2.5}, CO, O₃, and SO₂). The main caveat of the system is to prepare the initial conditions (ICs) 31 of the Community Multi-scale Air Quality (CMAQ) model simulations using observations 32 33 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 34 the Korea-United States Air Quality Study (KORUS-AQ) campaign period (1 May-12 June 35 2016). Data assimilation (DA) of optimal interpolation (OI) with Kalman filter was used in this 36 study. One major advantage of the system is that it can predict not only particulate matter (PM) 37 concentrations but also PM chemical composition including five main constituents: sulfate 38 (SO_4^{2-}) , nitrate (NO_3^{-}) , ammonium (NH_4^{+}) , organic aerosols (OAs), and elemental carbon (EC). 39 In addition, it is also capable of predicting the concentrations of gaseous pollutants (CO, O₃ 40 41 and SO_2). In this sense, this new air quality prediction system is comprehensive. The results 42 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 43 in terms of correlation, errors, and biases over the entire campaign period. The DA RUN agreed 44 reasonably well with the observations for PM_{10} (IOA = 0.60; MB = -13.54) and $PM_{2.5}$ (IOA = 45 0.71; MB = -2.43) as compared to the BASE RUN for PM_{10} (IOA = 0.51; MB = -27.18) and 46 $PM_{2.5}$ (IOA = 0.67; MB = -9.9). A significant improvement was also found with the DA RUN 47 in terms of bias. For example, for CO, the MB of -0.27 (BASE RUN) was greatly enhanced to 48 49 -0.036 (DA RUN). In the cases of O₃ and SO₂, the DA RUN also showed better performance than the BASE RUN. Further, several more practical issues frequently encountered in the air 50

51 quality prediction system were also discussed. In order to attain more accurate ozone predictions, the DA of NO₂ mixing ratios should be implemented with careful consideration of 52 the measurement artifacts (i.e., inclusion of alkyl nitrates, HNO₃, and PANs in the ground-53 54 observed NO₂ 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 55 layer heights (MLH) should be achieved from the meteorological modeling. Several 56 advantages of the current air quality prediction system, such as its non-static free parameter 57 58 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 59 60 system is not computationally expensive. In the ongoing and future works, more advanced DA 61 techniques such as the three-dimensional variational (3DVAR) method and ensemble Kalman filter (EnK) are being tested and will be introduced to the Korean air quality prediction system 62 (KAQPS). 63

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Keywords: Air quality prediction; Particulate matter (PM); Geostationary satellite sensor
 (GOCI); Air Korea; Data assimilation (DA); Dust episode predictions; NO₂ measurement
 artifacts

68 **1. Introduction**

Air quality has long been considered an important issue in climate change, visibility, 69 70 and public health, and it is strongly dependent upon meteorological conditions, emissions, and 71 the transport of air pollutants. Air pollutants typically consist of atmospheric particles and gases such as particulate matter (PM), carbon monoxide (CO), ozone (O₃), nitrogen dioxide (NO₂), 72 73 and sulfur dioxide (SO₂). These aerosols and gases play important roles in anthropogenic climate forcing both directly (Bellouin et al., 2005; Carmichael et al., 2009; IPCC, 2013; Scott 74 et al., 2014) and indirectly (Bréon et al., 2002; IPCC, 2013; Penner et al., 2004; Scott et al., 75 2014) in influencing the global radiation budget. Among the various air pollutants, PM and 76 surface O₃ are the most notorious health threats, as has been stated by several previous studies 77 (Carmichael et al., 2009; Dehghani et al., 2017; Khaniabadi et al., 2017). 78

79 With the stated importance of atmospheric aerosols and gases, considerable research 80 efforts have been made to monitor and quantify their amounts in the atmosphere through 81 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 82 real-time chemical concentrations as measured by ground-based observations for six criteria 83 84 air pollutants (PM10, PM2.5, O3, CO, SO2, and NO2) at the Air Korea website (https://www.airkorea.or.kr). In addition, the National Institute of Environmental Research 85 86 (NIER) of South Korea provides air quality predictions using multiple CTM simulations. Air 87 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 88 episodes. In this context, reliable and robust air quality forecasts are necessary to avoid any 89 90 confusion caused by poor predictions given by CTM simulations.

91 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 92 93 more accurate than model outputs, but they have spatial and temporal limitations. These limitations will be overcome by improving spatial and temporal coverage via future 94 geostationary satellite instruments such as the Geostationary Environment Monitoring 95 Spectrometer (GEMS) over Asia, the Tropospheric Emissions: Monitoring of Pollution 96 (TEMPO) over North America, and the Sentinel-4 over Europe. In addition, the TROPOspheric 97 98 Monitoring Instrument (TROPOMI) on board the Copernicus Sentinel-5 Precursor satellite was successfully launched into low earth orbit (LEO) on 13 October 2017 and are providing 99 100 information on the chemical composition in the atmosphere with a higher spatial resolution of $3.5 \times 7 \text{ km}^2$. 101

Unlike observation data, models can provide meteorological and chemical information 102 without any spatial and temporal data discontinuity, but they do have an issue of inaccuracy. 103 The major causes of uncertainty in the results of CTM simulations are introduced from 104 imperfect emissions, meteorological fields, initial conditions (ICs), and physical and chemical 105 106 parameterizations in the models (Carmichael et al., 2008). In order to minimize the limitations 107 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 108 109 chemical composition in the atmosphere by integrating observation data with model outputs 110 via data assimilation (DA) techniques.

111 Although the Korean numerical weather prediction (NWP) carried out by the Korea 112 Meteorological Administration (KMA) employs various DA techniques, almost no previous 113 efforts have been made to develop a air quality prediction system with DA in South Korea. Therefore, in the present study, the air quality 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, O₃, and SO₂. 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.

In this study, the optimal interpolation (OI) method with the Kalman filter was applied in order to develop the 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 air quality predictions such as: i) DA of NO₂ 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 air quality prediction system are discussed in Sect. 3, and then a summary and conclusions are given in Sect. 4.

137 **2. Methodology**

The 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.

144 **2.1 Meteorological and chemistry-transport modeling**

145 **2.1.1 WRF model simulations**

The WRF model has been developed for providing mesoscale numerical weather 146 prediction (NWP). It has also been used to provide meteorological input fields for CTM 147 simulations (Appel et al., 2010; Chemel et al., 2010; Foley et al., 2010; Lee et al., 2016; Park 148 et al., 2014). In this study, WRF v3.8.1 with the Advanced Research WRF (ARW) dynamical 149 core was applied to prepare the meteorological inputs for the CMAQ model simulations. 150 Dynamical and physical configurations for the WRF model simulations were selected as 151 follows: the Yonsei University (YSU) scheme for planetary boundary layer (Hong et al., 2006); 152 153 the WRF single-moment 6-class (WSM6) scheme for the micro-physics (Hong and Lim, 2006); the Grell-Freitas ensemble scheme for cumulus parameterization (Grell and Freitas, 2014); the 154 Noah-MP land surface model (Niu et al., 2011; Yang et al., 2011); the rapid radiative transfer 155 156 model for Global Circulation Models (RRTMG) for shortwave/longwave options (Iacono et al., 2008); and the revised MM5 scheme for surface layer options (Jiménez et al., 2012). The 157 National Centers for Environmental Prediction (NCEP) Final (FNL) Operational Global 158

Analysis data on $1^{\circ} \times 1^{\circ}$ grids were chosen for the ICs and boundary conditions (BCs) for the WRF simulations. In order to minimize meteorological field errors for the applications of ICs and BCs to the WRF simulations, the objective analysis (OBSGRID) nudging was conducted using the NCEP Automated Data Processing (ADP) global upper-air/surface observational weather data via the Cressman (1959)'s successive correction method. The adjusted meteorological variables were temperature, geopotential height, relative humidity, and zonal/meridional winds.

166 The model domain for the WRF simulations covers Northeast Asia with a horizontal resolution of 15×15 km², having a total of 223 latitudinal and 292 longitudinal grid cells. The 167 size of the WRF domain is slightly larger than that of the CMAQ domain, as shown in Fig. 1. 168 169 The meteorological data have 27 vertical layers from the surface (1000 hPa) to 50 hPa. The WRF meteorological fields (e.g., temperature, pressure, wind, humidity, cloud, etc) were then 170 transformed into the CMAQ-ready format via the Meteorology-Chemistry Interface Processor 171 (MCIP; Otte and Pleim (2010)) v4.3 which is a software to serve for transforming horizontal 172 and vertical coordinates while trying to maintain dynamic consistency between WRF and 173 174 CMAQ model simulations.

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176 **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 km² horizontal resolution and 27 sigma vertical layers. The CMAQv5.1 model was configured to use. Chemical and physical configurations for the CMAQ model simulations were selected as follows: SAPRC07tc for the

gas-phase chemical mechanism (Hutzell et al., 2012); AERO6 for aerosol thermodynamics 182 (Appel et al., 2013); Euler Backward Iterative (EBI) chemistry solver (Hertel et al., 1993), 183 which is a numerically optimized photochemistry mechanism solver; M3DRY for dry 184 185 deposition velocity (Pleim and Xiu, 2003; Xiu and Pleim, 2001); global mass-conserving scheme (YAMO & WRF) for horizontal and vertical advection (Colella and Woodward, 1984); 186 MULTISCALE (Louis, 1979), which is a simple first-order eddy diffusion scheme for 187 horizontal diffusion; and the Asymmetric Convective Model 2 (ACM2; Pleim, 2007a, 2007b) 188 189 for vertical diffusion.

For anthropogenic emissions, KORUS v1.0 emissions (Woo et al., 2012) were used. 190 191 The KORUS v1.0 emissions cover almost all of Asia, and are based on three emission 192 inventories: the Comprehensive Regional Emissions inventory for Atmospheric Transport Experiment (CREATE) for East Asia excluding Japan; the Model Inter-Comparison Study for 193 Asia (MICS-Asia) for Japan; and the Studies of Emissions and Atmospheric Composition, 194 Clouds and Climate Coupling by Regional Surveys (SEAC4RS) for South and Southeast Asia. 195 Biogenic emissions were prepared by running the Model of Emissions of Gases and 196 197 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 198 data (Friedl et al., 2010) and improved leaf area index (LAI) based on MODIS datasets (Yuan 199 200 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 201 simulations were prepared using the global model results of the Model for Ozone and Related 202 203 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 204

205 information.

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207 **2.2 Observation data**

208 2.2.1. Satellite-based observations

A Korean geostationary satellite of Communication, Ocean, and Meteorological 209 210 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 211 latitude of 36°N and a longitude of 128.2°E with a horizontal coverage of $2500 \times 2500 \text{ km}^2$ 212 (refer to Fig. 1). Among the three payloads of the COMS, Geostationary Ocean Color Image 213 214 (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 500×500 215 m^2 from 00:30 to 07:30 Coordinated Universal Time (UTC) for eight spectral (6 visible and 2 216 217 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 218 219 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 220 (Lee et al., 2012). Choi et al. (2016) further extended the algorithm for application to land 221 surfaces, and the algorithm was referred to as the GOCI YAER version 1 algorithm. With the 222 223 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 224 225 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 226 YAER algorithm (Choi et al., 2018), which used the updated cloud masking and more accurate 227 228 surface reflectances. In this study, the most recent GOCI AOD products from the GOCI YAER

- 230
- 231 2.2.2. Ground-based observations

version 2 algorithm were used.

In addition to the satellite data, ground-based observations in South Korea and China were also collected for use in the 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 PM₁₀, PM_{2.5}, CO, O₃, SO₂, and NO₂.

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 qualityassured level 2.0 AODs for the AERONET were used in this study.

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248 2.3 Air quality prediction system

In the present study, the air quality prediction system was developed by adjusting the ICs for the CMAQ model simulations based on DA with satellite-retrieved and groundmeasured 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.
256
257 2.3.1. AOD calculations

258 CMAQ AODs are calculated by integrating the aerosol extinction coefficient (σ_{ext}) 259 using the following equation:

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261 $AOD(\lambda) = \int_{0}^{z} \sigma_{ext}(\lambda) dz$ (1)

262

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:

266

267

 $\sigma_{\rm ext}(\lambda) = \sigma_{\rm abs}(\lambda) + \sigma_{\rm sca}(\lambda)$

268
$$\sigma_{abs}(\lambda) \left[Mm^{-1} \right] = \sum_{i}^{n} \sum_{j}^{m} \left\{ (1 - \omega_{ij}(\lambda)) \cdot \beta_{ij}(\lambda) \cdot f_{ij}(RH) \cdot [C]_{ij} \right\}$$
(3)

(2)

269
$$\sigma_{sca}(\lambda) \, [Mm^{-1}] = \sum_{i}^{n} \sum_{j}^{m} \{ \omega_{ij}(\lambda) \cdot \beta_{ij}(\lambda) \cdot f_{ij}(RH) \cdot [C]_{ij} \}$$
(4)

270

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; [C]_{ij} is the concentration of particulate species including (NH₄)₂SO₄, NH₄NO₃, black carbon, organic aerosols (OA), mineral dust, and sea275 salt aerosols; RH is the relative humidity; $f_{ii}(RH)$ is the hygroscopic factor; and the single scattering albedo (ω) implies to the fraction (portion) of scattering in the total extinction. 276 Using Eqns. (2) - (4), AODs were calculated from the aerosol composition and RH. 277 These have been intensive tests using different β and f(RH) values in the following three 278 previous studies: (1) Chin et al. (2002)'s study with the Goddard Chemistry Aerosol Radiation 279 and Transport (GOCART) model; (2) Martin et al. (2003)'s study with the GEOS-Chem model; 280 and (3) Malm and Hand (2007)'s study with the CMAQ model. Lee et al. (2016) tested these 281 282 methods, and then found that Chin et al. (2002)'s method reproduced the best results in estimating AODs at 550 nm over East Asia. On the basis of Lee et al. (2016)'s work, σ_{ext} was 283 estimated with the β and f(RH) values suggested by Chin et al. (2002). After that, σ_{ext} was 284 integrated with respect to altitude, in order to calculate the AODs. The calculated AODs were 285 used in the air quality prediction system in order to prepare the ICs for the PM predictions. 286

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288 2.3.2. Data assimilation (DA)

The ground-based observations, together with GOCI-derived AODs, were used to 289 prepare the ICs for the air quality predictions with the CMAQ model simulations. In order to 290 achieve this, the following steps were taken: (i) the CMAQ-calculated concentrations of CO, 291 O₃, and SO₂ were combined with the concentrations of CO, O₃, and SO₂ obtained from ground-292 based observations in South Korea (Air Korea) and China; (ii) the CMAQ-calculated AODs 293 were assimilated with the GOCI AODs; (iii) the assimilated AODs were converted into PM₁₀; 294 295 (iv) the converted PM₁₀ 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 296 297 CMAQ-simulated concentrations were applied so as to the adjust vertical profiles of the chemical species above the surface. In the air quality 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 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; Park et al., 2014; Tang et al., 2015, 2017).

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

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310
$$\tau'_{\rm m} = \tau_{\rm m} + \mathbf{K}(\tau_{\rm o} - \mathbf{H}\tau_{\rm m}) \tag{5}$$

311
$$\mathbf{K} = \mathbf{B}\mathbf{H}^{\mathrm{T}}(\mathbf{H}\mathbf{B}\mathbf{H}^{\mathrm{T}} + \mathbf{0})^{-1}$$
(6)

312
$$\mathbf{0} = [(\mathbf{f}_0 \boldsymbol{\tau}_0)^2 + (\boldsymbol{\varepsilon}_0)^2]\mathbf{I}$$
(7)

313
$$\mathbf{B}(d_{x}, d_{z}) = [(f_{m}\tau_{m})^{2} + (\varepsilon_{m})^{2}] \exp\left[-\frac{d_{x}^{2}}{2l_{mx}^{2}}\right] \exp\left[-\frac{d_{z}^{2}}{2l_{mz}^{2}}\right]$$
(8)

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where τ'_{m} , τ_{m} , and τ_{o} represent the assimilated products by the OI method, the modeled values, and the observed values, respectively; **K** is the Kalman gain matrix; **H** is the observation operator (or forward operator), which is an interpolator from model to observation space; **B** and **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; **I** denotes the unit matrix; 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 (fm, fo, ϵ_m , ϵ_o , l_{mx} , and l_{mz}) were used to calculate the error 327 covariance matrices of the observations and model, the mathematical formalisms of which are 328 described in Eq. (7) and (8), respectively. Several previous studies have used fixed values for 329 330 free parameters (Collins et al., 2001; Yu et al., 2003; Adhikary et al., 2008; Chung et al., 2010). These runs are called "static" runs. In contrast to those previous studies, "non-static" free 331 parameters were applied in this study by minimizing the differences between the assimilated 332 values and observations via an iterative process at each assimilation time step. This non-static 333 334 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 335 4-D variational methods. This is another advantage of using the OI technique in this system. It 336 typically takes less than 20 minutes with a workstation environment (dual Intel Xeon 2.40 GHz 337 processor). 338

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340 2.3.3. Allocation of the assimilated PM₁₀ & PM_{2.5} into particulate composition

In the procedure of 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, 344 $\Delta PM_{2.5}$ was estimated using the ratios of $PM_{2.5}$ to PM_{10} from the background CMAQ model 345 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 346 according to the comparison between two PM_{2.5} compositions observed at the seven super-sites 347 and simulated from the CMAQ model runs over South Korea. Both of the compositions are 348 shown in Fig. 4. In Fig. 4, "PM OTHERS" indicates the remaining particulate matter species 349 350 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 351 fraction, $\Delta PM_{10} \times (1-PM_{2.5}/PM_{10})$, was also distributed into the coarse-mode particles (PM_{2.5-10}) 352 as crustal elements. 353

354

355 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.

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361 **3.1. Evaluation of the air quality prediction system**

362 3.1.1. Time-series analysis

Figure 5 shows the time-series plots of PM_{10} , $PM_{2.5}$, CO, O₃, and SO₂ 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

367 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 00:00 368 UTC. For the forecast hours from 01:00 to 23:00 UTC, all of the ground observations (254 Air 369 370 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 371 prediction performances by applying the ICs to the CMAQ model simulations. The BASE 372 RUN significantly under-predicted PM₁₀, PM_{2.5}, and CO while the DA RUN produced 373 374 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 375 from the BASE RUN. These large differences are well known, and have been attributed to the 376 377 underestimated emissions of CO (Heald et al., 2004). However, when the DA was applied, the predictions of the CO mixing ratios improved. Similarly, the performances of the PM_{10} and 378 PM_{2.5} predictions improved with the application of the DA. Unlike PM₁₀, PM_{2.5}, and CO, the 379 O3 mixing ratios and its diurnal trends from both the BASE RUN and DA RUN tend to be well-380 matched with the observations. By contrast, the poorest performances of the BASE RUN and 381 382 the DA RUN were shown for SO₂.

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 SO₂ 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 from the DA RUN by assimilating CMAQ outputs 394 with Air Korea-observed (a) O₃ mixing ratios, and (b) both O₃ and NO₂ mixing ratios for a 395 396 preliminary test run. 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 397 09:00 UTC (13:00 and 18:00 KST), when solar insolation is the most intense. This may be 398 399 attributed to the chemical imbalances between ozone production and ozone destruction (or titration). However, if CMAQ NO₂ was assimilated with ground-based observations in South 400 Korea (Air Korea) and China, the predicted ozone mixing ratios became substantially closer to 401 the observations, as shown in Fig. 7(b). This is clearly due to the fact that NO_x is an important 402 precursor of ozone. In the prediction of the ozone mixing ratios, both 1-hr peak ozone (around 403 404 15:00 KST) and 8-hr averaged ozone mixing ratios (between 9:00 and 17:00 KST) are 405 important. Fig. 7 clearly shows that the prediction accuracies of both the ozone mixing ratios were improved after the DA of NO₂ mixing ratios. 406

407 Although the DA for NO₂ provided better ozone predictions, one should take caution 408 in using the NO₂ observations. The NO₂ mixing ratios measured at Air Korea sites are known 409 to be contaminated by other nitrogen gases such as nitric acid (HNO₃), peroxyacetyl nitrates 410 (PANs), and alkyl nitrates (ANs), since the Air Korea NO₂ mixing ratios are measured through 411 a chemiluminescent method with catalysts of gold or molybdenum oxide at high temperatures. 412 These are known to be "NO₂ measurement artifacts" (Jung et al., 2017), which is one of the reasons that the DA of NO₂ was not shown in Fig. 6. The NO₂ mixing ratios are corrected from 413 the Air Korea NO₂ data, and are then used to prepare the ICs via the DA for more accurate 414 415 ozone and NO₂ predictions. Currently, such corrections of the observed NO₂ mixing ratios are being standardized with more sophisticated year-long NO₂ measurements. After the corrections 416 of the NO₂ measurement artifacts, more evolved schemes of ozone and NO₂ predictions will 417 be possible in the future. As shown in Fig. 7, about a 20% reduction (average fraction of non-418 419 NO₂ mixing ratios in the observed NO₂ mixing ratios) was made for these demonstration runs (Jung et al., 2017). 420

Another practical issue is now discussed. Although the assimilation with the observed 421 422 NO₂ 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 423 aggregated plot of the ozone mixing ratios at the observation sites (see Fig. 7). This can be 424 caused by underestimated NO₂ mixing ratios and thus not enough nighttime ozone titration. As 425 aforementioned, reliable NO₂ prediction via the correction of the NO₂ measurement artifacts 426 427 will be made in the future. Another possible reason of the over-predicted ozone mixing ratios during the nighttime can be underestimation of the mixing layer height (MLH). Figure 8 shows 428 a comparison between lidar-measured MLH (black dashed line) and WRF-calculated MLH 429 430 (with the option of the Yonsei University (YSU) planetary boundary layer scheme by Hong et al. (2006); see red line). As shown in Fig. 8, the nocturnal lidar-measured MLH is about two 431 times higher than the nocturnal WRF-calculated MLH as measured at a lidar site inside the 432 433 campus of Seoul National University (SNU) in Seoul. Such underestimated MLH in the model 434 tends to compress the ozone molecules within the mixing layer during the nighttime, which

leads to consistently over-predicted nocturnal ozone mixing ratios. Based on this discrepancy
shown in Fig. 8, more intensive comparison study is being carried out by comparing lidarmeasured MLH with model-calculated MLH at multiple sites in South Korea.

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, a strong capability of our DA system is to improve the predictions of the aerosol composition.

443

444 **3.1.2. Spatial distribution**

445 Figure 10 shows the spatial distributions and bias of PM and chemical species throughout the entire period of the KORUS-AQ campaign over the Seoul Metropolitan Area 446 (SMA). Noticeable improvements are observed to have been achieved in the spatial 447 distributions by applying the ICs into the CMAQ model simulations, particularly for PM₁₀ (Fig. 448 10a), PM_{2.5} (Fig. 10b), and CO (Fig. 10c). As shown in Fig. 10, the under-predicted 449 450 concentrations of PM10, PM2.5, and CO were adjusted to concentrations closer to the observations. In case of SO₂ (see Fig. 10d), the DA RUN produced better agreement with the 451 observations than the BASE RUN, but there were still under-predicted SO₂ concentrations over 452 453 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 458 excluded in the preparation of the ICs for CMAQ model simulations. Again, this is because the 459 Air Korea NO_2 mixing ratios are contaminated by several reactive nitrogen species, so the data 460 cannot be directly used in the assimilation procedures. In case of VOCs, a limited number of 461 datasets is available in South Korea for the DA. Improvements in the prediction of ozone 462 mixing ratios can be achieved when the NO_2 mixing ratios are corrected and a sufficient 463 number of VOCs data (possibly from satellite data in the future) is available.

464

465 **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 coefficient (R), root mean square error (RMSE), and mean bias (MB) were conducted using observations from the Air Korea stations for PM₁₀, PM_{2.5}, CO, SO₂, and O₃ (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 471 correlation coefficient became higher for the entire predictions. In addition, it was found that 472 473 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 474 DA RUN were reasonably consistent with the observations for PM_{10} (IOA = 0.60; R= 0.40; 475 RMSE = 34.87; MB = -13.54) and PM_{2.5} (IOA = 0.71; R= 0.53; RMSE = 17.83; MB = -2.43), 476 as compared to the BASE RUN for PM_{10} (IOA = 0.51; R= 0.34; RMSE = 40.84; MB = -27.18) 477 and $PM_{2.5}$ (IOA = 0.67; R= 0.51; RMSE = 19.24; MB = -9.9). In terms of bias, an improvement 478 479 was found for CO: MB = -0.036 for the DA RUN and MB = -0.27 for the BASE RUN. Regarding O₃ and SO₂, the DA RUN showed slightly better performances than the BASE RUN. 480

481	Table 2 presents the results of the statistical analysis at 00:00 UTC when the DA was
482	conducted, with the results clearly showing how much closer the DA makes the CMAQ-
483	calculated chemical concentrations to the observed concentrations. Collectively, the DA
484	improved model accuracy by a large degree in terms of R, particularly for PM_{10} (R: 0.3 \rightarrow 0.75;
485	slope: 0.17 \rightarrow 0.66) and O ₃ (R: 0.09 \rightarrow 0.61; slope: 0.07 \rightarrow 0.42). In addition, for all species, MB
486	and RMSE decreased significantly with the DA RUN as compared with the BASE RUN.

488 **3.2. Sensitivity test of DA time-interval**

489 **3.2.1. AOD**

In this section, a sensitivity analysis was conducted with different implementation 490 time-intervals of the DA (i.e., 24, 6, and 3 hours) for AOD (refer to Fig. 12). As shown in Fig. 491 12, more frequent implementation of the DA is expected to make the predicted results closer 492 to the observations. Although the DA RUN with a shorter assimilation time-interval tends to 493 produce a better prediction, it is not always the most appropriate choice, since the shorter 494 assimilation time-interval results in increased computational cost. Therefore, an optimized 495 assimilation time-interval should be found to achieve the best performances from the given DA 496 497 system with the consideration of its own computational ability.

498

499 **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) SO₂, and (e) O₃, 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 24 504 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 505 506 to improve if the DAs are implemented multiple times prior to the actual predictions at 00:00 507 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 00:00 508 UTC. The performances of all of the chemical species excluding ozone improved, as expected, 509 with DA RUNs with more frequent and longer DA time-intervals (i.e., three-times 510 511 implementation with a 2-hr time-interval in our cases). In case of ozone, the best performance was found for the air quality prediction system with the DA time-interval of 24-hr. 512

513 Unsurprisingly, more frequent DAs prior to the actual prediction mode (i.e., before 514 00:00 UTC in our system) with a longer time-interval (such as 2-hr) will be computationally 515 costly. There will certainly be a "trade-off" between the precision of air quality prediction and 516 the computational cost. The system should be designed under the consideration of these two 517 factors.

518

519 4. Summary and conclusions

In this study, the 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 SO₂ 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.

526 The performances of the developed prediction system were evaluated using nearsurface in-situ observation data. The CMAQ model runs with the ICs (DA RUN) showed higher 527 consistency with the observations of almost all of the chemical species, including PM 528 composition (sulfate, nitrate, ammonium, OA, and EC) and atmospheric gases (CO, ozone, and 529 SO₂), than the CMAQ model runs without the ICs (BASE RUN). Particularly for CO, the DA 530 was able to remarkably improve the model performances, while the BASE RUN significantly 531 under-predicted the CO concentrations (predicting about one-third of the observed values). In 532 533 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 534 NO₂ mixing ratios and the corrections of model-simulated mixing layer height (MLH). For SO₂, 535 536 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 537 these can be estimated through inverse modeling using satellite data (e.g., Lee et al., 2011). 538 The adjustments of both ICs and emissions may be able to improve the performances of the air 539 quality prediction system, and this will be examined in future studies. 540

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 $7 \times 8 \text{ km}^2$ 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 548 (AI)-based air quality prediction systems are also currently being developed in several ways 549 (e.g., Kim et al., 2019). Actually, Kim et al. (2019) developed an AI-based PM prediction 550 551 system based on a deep recurrent neural network (RNN) in South Korea. The AI-based prediction system was optimized by iterative model trainings with the inputs of ground-552 553 observed PM₁₀, PM_{2.5}, and meteorological fields including wind speed, wind direction, relative humidity, and precipitation. The AI-based prediction system showed better performances at the 554 555 several sites than the CMAQ model simulations. However, it works only for the observation sites in South Korea where ground-based observations are available. By taking advantages of 556 557 both the CTM-based air quality prediction and the AI-based prediction systems, both systems 558 will be eventually combined so as to create a more accurate hybrid air quality prediction system over South Korea. This will be the ultimate goal of the series of our research works. 559

560

Code and data availability. WRF v3.8.1 (doi:10.5065/D6MK6B4K) and CMAQ v5.1 561 (doi:10.5281/zenodo.1079909) models are both open-source and publicly available. Source 562 563 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-564 565 AQ field campaign can be downloaded from the KORUS-AQ data archive (http://www-566 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 567 South Korea and https://pm25.in for China. AERONET data were downloaded from 568 569 https://aeronet.gsfc.nasa.gov. All codes related with the air quality prediction system can be obtained by contacting K. Lee (lkh1515@gmail.com). NCL (2019; doi:10.5065/D6WD3XH5)
was used to draw the figures.

572

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.

578

579 Acknowledgments

This research was supported by the National Strategic Project-Fine particle of the National 580 581 Research Foundation of Korea (NRF) of the Ministry of Science and ICT (MSIT), the Ministry of Environment (MOE), and the Ministry of Health and Welfare (MOHW) (NRF-582 2017M3D8A1092022). This work was also funded by the MOE as "Public Technology 583 Program based on Environmental Policy (2017000160001)" and was supported by a grant from 584 the National Institute of Environment Research (NIER), funded by the MOE of the Republic 585 of Korea (NIER-2019-01-01-028). Specially thanks to the entire KORUS-AQ science team for 586 their considerable efforts in conducting the campaign. 587

APPENDIX A: FORMULAS FOR STATISTICAL EVALUATION INDICES

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

592 Index Of Agreement (IOA) =
$$1 - \frac{\sum_{1}^{n} (M - \overline{0})^{2}}{\sum_{1}^{n} (|M - \overline{0}| + |0 - \overline{0}|)^{2}}$$
 (A1)

594 Correlation Coefficient (R) =
$$\frac{1}{(n-1)} \sum_{1}^{n} \left(\left(\frac{0-\overline{0}}{\sigma_{0}} \right) \left(\frac{M-\overline{M}}{\sigma_{m}} \right) \right)$$
 (A2)

596 Root Mean Square Error (RMSE) =
$$\sqrt{\frac{\sum_{1}^{n}(M-0)^{2}}{n}}$$
 (A3)

598 Mean Bias (MB) =
$$\frac{1}{n} \sum_{1}^{n} (M - 0)$$
 (A4)

600 Mean Normalized Bias (MNB) =
$$\frac{1}{n} \sum_{n=1}^{n} \left(\frac{M-0}{0}\right) \times 100 \%$$
 (A5)

602 Mean Normalized Error (MNE) =
$$\frac{1}{n} \sum_{1}^{n} \left(\frac{|M-0|}{0}\right) \times 100 \%$$
 (A6)

604 Mean Fractional Bias (MFB) =
$$\frac{1}{n} \sum_{n=1}^{n} \frac{(M-0)}{\left(\frac{M+0}{2}\right)} \times 100 \%$$
 (A7)

....

606 Mean Fractional Error (MFE) =
$$\frac{1}{n} \sum_{n=1}^{n} \frac{|M-0|}{\left(\frac{M+0}{2}\right)} \times 100 \%$$
 (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.

613 **References**

- Adhikary, B., Kulkarni, S., Dallura, A., Tang, Y., Chai, T., Leung, L. R., Qian, Y., Chung, C. E.,
 Ramanathan, V. and Carmichael, G. R.: A regional scale chemical transport modeling of
 Asian aerosols with data assimilation of AOD observations using optimal interpolation
 technique, Atmospheric Environment, 42(37), 8600–8615,
 doi:10.1016/j.atmosenv.2008.08.031, 2008.
- Appel, K. W., Roselle, S. J., Gilliam, R. C. and Pleim, J. E.: Sensitivity of the Community
 Multiscale Air Quality (CMAQ) model v4.7 results for the eastern United States to MM5
 and WRF meteorological drivers, Geoscientific Model Development, 3, 169–188, 2010.
- Appel, K. W., Pouliot, G. A., Simon, H., Sarwar, G., Pye, H. O. T., Napelenok, S. L., Akhtar,
 F. and Roselle, S. J.: Evaluation of dust and trace metal estimates from the Community
 Multiscale Air Quality (CMAQ) model version 5.0, Geoscientific Model Development,
 6(4), 883–899, doi:10.5194/gmd-6-883-2013, 2013.
- Bellouin, N., Boucher, O., Haywood, J. and Reddy, M. S.: Global estimate of aerosol direct
 radiative forcing from satellite measurements, Nature, 438(7071), 1138–1141,
 doi:10.1038/nature04348, 2005.
- Bréon, F.-M., Tanré, D. and Generoso, S.: Aerosol Effect on Cloud Droplet Size Monitored
 from Satellite, Science, 295(5556), 834–838, doi:10.1126/science.1066434, 2002.
- Byun, D. and Schere, K. L.: Review of the Governing Equations, Computational Algorithms,
 and Other Components of the Models-3 Community Multiscale Air Quality (CMAQ)
 Modeling System, Appl. Mech. Rev, 59(2), 51–77, doi:10.1115/1.2128636, 2006.
- Byun, D. W. and Ching, J. K. S.: Science algorithms of the EPA models-3 community
 multiscale air quality (CMAQ) modeling system, U.S. Environmental Protection Agency,
 EPA/600/R-99/030 (NTIS PB2000-100561)., 1999.
- Carmichael, G. R., Sakurai, T., Streets, D., Hozumi, Y., Ueda, H., Park, S. U., Fung, C., Han,
 Z., Kajino, M., Engardt, M., Bennet, C., Hayami, H., Sartelet, K., Holloway, T., Wang, Z.,
 Kannari, A., Fu, J., Matsuda, K., Thongboonchoo, N. and Amann, M.: MICS-Asia II: The
 model intercomparison study for Asia Phase II methodology and overview of findings,
 Atmospheric Environment, 42(15), 3468–3490, doi:10.1016/j.atmosenv.2007.04.007,
 2008.
- Carmichael, G. R., Adhikary, B., Kulkarni, S., D'Allura, A., Tang, Y., Streets, D., Zhang, Q.,
 Bond, T. C., Ramanathan, V., Jamroensan, A. and Marrapu, P.: Asian Aerosols: Current
 and Year 2030 Distributions and Implications to Human Health and Regional Climate
 Change, Environ. Sci. Technol., 43(15), 5811–5817, doi:10.1021/es8036803, 2009.
- Chemel, C., Sokhi, R. S., Yu, Y., Hayman, G. D., Vincent, K. J., Dore, A. J., Tang, Y. S., Prain,
 H. D. and Fisher, B. E. A.: Evaluation of a CMAQ simulation at high resolution over the
 UK for the calendar year 2003, Atmospheric Environment, 44(24), 2927–2939,
 doi:10.1016/j.atmosenv.2010.03.029, 2010.

- Chin, M., Ginoux, P., Kinne, S., Torres, O., Holben, B. N., Duncan, B. N., Martin, R. V., Logan,
 J. A., Higurashi, A. and Nakajima, T.: Tropospheric Aerosol Optical Thickness from the
 GOCART Model and Comparisons with Satellite and Sun Photometer Measurements, J.
 Atmos. Sci., 59(3), 461–483, doi:10.1175/15200469(2002)059<0461:TAOTFT>2.0.CO;2, 2002.
- Choi, M., Kim, J., Lee, J., Kim, M., Park, Y.-J., Jeong, U., Kim, W., Hong, H., Holben, B., Eck,
 T. F., Song, C. H., Lim, J.-H. and Song, C.-K.: GOCI Yonsei Aerosol Retrieval (YAER)
 algorithm and validation during the DRAGON-NE Asia 2012 campaign, Atmos. Meas.
 Tech., 9(3), 1377–1398, doi:10.5194/amt-9-1377-2016, 2016.
- Choi, M., Kim, J., Lee, J., Kim, M., Park, Y.-J., Holben, B., Eck, T. F., Li, Z. and Song, C. H.:
 GOCI Yonsei aerosol retrieval version 2 products: an improved algorithm and error
 analysis with uncertainty estimation from 5-year validation over East Asia, Atmos. Meas.
 Tech., 11(1), 385–408, doi:10.5194/amt-11-385-2018, 2018.
- Chung, C. E., Ramanathan, V., Carmichael, G., Kulkarni, S., Tang, Y., Adhikary, B., Leung, L.
 R. and Qian, Y.: Anthropogenic aerosol radiative forcing in Asia derived from regional models with atmospheric and aerosol data assimilation, Atmos. Chem. Phys., 10(13), 6007–6024, doi:10.5194/acp-10-6007-2010, 2010.
- Colella, P. and Woodward, P. R.: The Piecewise Parabolic Method (PPM) for gas-dynamical
 simulations, Journal of Computational Physics, 54(1), 174–201, doi:10.1016/00219991(84)90143-8, 1984.
- Collins, W. D., Rasch, P. J., Eaton, B. E., Khattatov, B. V., Lamarque, J.-F. and Zender, C. S.:
 Simulating aerosols using a chemical transport model with assimilation of satellite aerosol
 retrievals: Methodology for INDOEX, J. Geophys. Res., 106(D7), 7313–7336,
 doi:10.1029/2000JD900507, 2001.
- Cressman, G. P.: An operational objective analysis system, Mon. Wea. Rev., 87(10), 367–374,
 1959.
- Dehghani, M., Keshtgar, L., Javaheri, M. R., Derakhshan, Z., Conti, O., Gea, Zuccarello, P. and
 Ferrante, M.: The effects of air pollutants on the mortality rate of lung cancer and leukemia,
 Molecular Medicine Reports, 15(5), 3390–3397, 2017.
- Emmons, L. K., Walters, S., Hess, P. G., Lamarque, J.-F., Pfister, G. G., Fillmore, D., Granier,
 C., Guenther, A., Kinnison, D., Laepple, T., Orlando, J., Tie, X., Tyndall, G., Wiedinmyer,
 C., Baughcum, S. L. and Kloster, S.: Description and evaluation of the Model for Ozone
 and Related chemical Tracers, version 4 (MOZART-4), Geosci. Model Dev., 3(1), 43–67,
 doi:10.5194/gmd-3-43-2010, 2010.
- Foley, K. M., Roselle, S. J., Appel, K. W., Bhave, P. V., Pleim, J. E., Otte, T. L., Mathur, R.,
 Sarwar, G., Young, J. O., Gilliam, R. C., Nolte, C. G., Kelly, J. T., Gilliland, A. B. and
 Bash, J. O.: Incremental testing of the Community Multiscale Air Quality (CMAQ)
 modeling system version 4.7, Geosci. Model Dev., 3(1), 205–226, doi:10.5194/gmd-3-

689 205-2010, 2010.

Friedl, M. A., Sulla-Menashe, D., Tan, B., Schneider, A., Ramankutty, N., Sibley, A. and Huang,
X.: MODIS Collection 5 global land cover: Algorithm refinements and characterization
of new datasets, Remote Sensing of Environment, 114(1), 168–182,
doi:10.1016/j.rse.2009.08.016, 2010.

- Generoso, S., Bréon, F.-M., Chevallier, F., Balkanski, Y., Schulz, M. and Bey, I.: Assimilation
 of POLDER aerosol optical thickness into the LMDz-INCA model: Implications for the
 Arctic aerosol burden, J. Geophys. Res., 112(D2), D02311, doi:10.1029/2005JD006954,
 2007.
- Grell, G. A. and Freitas, S. R.: A scale and aerosol aware stochastic convective parameterization
 for weather and air quality modeling, Atmospheric Chemistry and Physics, 14(10), 5233–
 5250, doi:https://doi.org/10.5194/acp-14-5233-2014, 2014.
- Guenther, A., Karl, T., Harley, P., Wiedinmyer, C., Palmer, P. I. and Geron, C.: Estimates of
 global terrestrial isoprene emissions using MEGAN (Model of Emissions of Gases and
 Aerosols from Nature), Atmospheric Chemistry and Physics, 6(11), 3181–3210, 2006.
- Guenther, A. B., Jiang, X., Heald, C. L., Sakulyanontvittaya, T., Duhl, T., Emmons, L. K. and
 Wang, X.: The Model of Emissions of Gases and Aerosols from Nature version 2.1
 (MEGAN2.1): an extended and updated framework for modeling biogenic emissions,
 Geosci. Model Dev., 5(6), 1471–1492, doi:10.5194/gmd-5-1471-2012, 2012.
- Heald, C. L., Jacob, D. J., Jones, D. B. A., Palmer, P. I., Logan, J. A., Streets, D. G., Sachse, G.
 W., Gille, J. C., Hoffman, R. N. and Nehrkorn, T.: Comparative inverse analysis of satellite
 (MOPITT) and aircraft (TRACE-P) observations to estimate Asian sources of carbon
 monoxide: COMPARATIVE INVERSE ANALYSIS, Journal of Geophysical Research:
 Atmospheres, 109(D23), doi:10.1029/2004JD005185, 2004.
- Hertel, O., Berkowicz, R., Christensen, J. and Hov, Ø.: Test of two numerical schemes for use
 in atmospheric transport-chemistry models, Atmospheric Environment. Part A. General
 Topics, 27(16), 2591–2611, doi:10.1016/0960-1686(93)90032-T, 1993.
- Holben, B. N., Eck, T. F., Slutsker, I., Tanré, D., Buis, J. P., Setzer, A., Vermote, E., Reagan, J.
 A., Kaufman, Y. J., Nakajima, T., Lavenu, F., Jankowiak, I. and Smirnov, A.:
 AERONET—A Federated Instrument Network and Data Archive for Aerosol
 Characterization, Remote Sensing of Environment, 66(1), 1–16, doi:10.1016/S00344257(98)00031-5, 1998.
- Hong, S.-Y. and Lim, J.-O. J.: The WRF single-moment 6-class microphysics scheme (WSM6),
 J. Korean Meteor. Soc., 42(2), 129–151, 2006.
- Hong, S.-Y., Noh, Y. and Dudhia, J.: A New Vertical Diffusion Package with an Explicit
 Treatment of Entrainment Processes, Mon. Wea. Rev., 134(9), 2318–2341,
 doi:10.1175/MWR3199.1, 2006.

- Hutzell, W. T., Luecken, D. J., Appel, K. W. and Carter, W. P. L.: Interpreting predictions from
 the SAPRC07 mechanism based on regional and continental simulations, Atmospheric
 Environment, 46, 417–429, doi:10.1016/j.atmosenv.2011.09.030, 2012.
- Iacono, M. J., Delamere, J. S., Mlawer, E. J., Shephard, M. W., Clough, S. A. and Collins, W.
 D.: Radiative forcing by long-lived greenhouse gases: Calculations with the AER radiative
 transfer models, J. Geophys. Res., 113(D13), D13103, doi:10.1029/2008JD009944, 2008.
- IPCC: Climate Change 2013: The Physical Science Basis. The Fifth Assessment Report of the
 Intergovernmental Panel on Climate Change, , Cambridge University Press, Cambridge,
 United Kingdom and New York, NY, USA, 2013.
- Jiménez, P. A., Dudhia, J., González-Rouco, J. F., Navarro, J., Montávez, J. P. and GarcíaBustamante, E.: A Revised Scheme for the WRF Surface Layer Formulation, Monthly
 Weather Review, 140(3), 898–918, 2012.
- Jung, J., Lee, J., Kim, B. and Oh, S.: Seasonal variations in the NO2 artifact from
 chemiluminescence measurements with a molybdenum converter at a suburban site in
 Korea (downwind of the Asian continental outflow) during 2015–2016, Atmospheric
 Environment, 165, 290–300, doi:10.1016/j.atmosenv.2017.07.010, 2017.
- Khaniabadi, Y. O., Goudarzi, G., Daryanoosh, S. M., Borgini, A., Tittarelli, A. and De Marco,
 A.: Exposure to PM10, NO2, and O3 and impacts on human health, Environmental
 Science and Pollution Research, 24(3), 2781–2789, doi:10.1007/s11356-016-8038-6,
 2017.
- Khattatov, B. V., Gille, J. C., Lyjak, L. V., Brasseur, G. P., Dvortsov, V. L., Roche, A. E. and
 Waters, J. W.: Assimilation of photochemically active species and a case analysis of
 UARS data, J. Geophys. Res., 104(D15), 18715–18737, doi:10.1029/1999JD900225,
 1999.
- Khattatov, B. V., Lamarque, J.-F., Lyjak, L. V., Menard, R., Levelt, P., Tie, X., Brasseur, G. P.
 and Gille, J. C.: Assimilation of satellite observations of long-lived chemical species in
 global chemistry transport models, J. Geophys. Res., 105(D23), 29135–29144,
 doi:10.1029/2000JD900466, 2000.
- Kim, H. S., Park, I., Song, C. H., Lee, K., Yun, J. W., Kim, H. K., Jeon, M., Lee, J. and Han,
 K. M.: Development of a daily PM10 and PM2.5 prediction system using a deep long
 short-term memory neural network model, Atmos. Chem. Phys., 19(20), 12935–12951,
 doi:10.5194/acp-19-12935-2019, 2019.
- Lamarque, J.-F., Khattatov, B. V., Gille, J. C. and Brasseur, G. P.: Assimilation of Measurement
 of Air Pollution from Space (MAPS) CO in a global three-dimensional model, J. Geophys.
 Res., 104(D21), 26209–26218, doi:10.1029/1999JD900807, 1999.
- Lee, C., Martin, R. V., Donkelaar, A. van, Lee, H., Dickerson, R. R., Hains, J. C., Krotkov, N.,
 Richter, A., Vinnikov, K. and Schwab, J. J.: SO2 emissions and lifetimes: Estimates from

- inverse modeling using in situ and global, space-based (SCIAMACHY and OMI)
 observations, Journal of Geophysical Research: Atmospheres, 116(D6),
 doi:10.1029/2010JD014758, 2011.
- Lee, J., Kim, J., Song, C. H., Ryu, J.-H., Ahn, Y.-H. and Song, C. K.: Algorithm for retrieval of
 aerosol optical properties over the ocean from the Geostationary Ocean Color Imager,
 Remote Sensing of Environment, 114(5), 1077–1088, doi:10.1016/j.rse.2009.12.021,
 2010.
- Lee, J., Kim, J., Yang, P. and Hsu, N. C.: Improvement of aerosol optical depth retrieval from MODIS spectral reflectance over the global ocean using new aerosol models archived from AERONET inversion data and tri-axial ellipsoidal dust database, Atmos. Chem. Phys., 12(15), 7087–7102, doi:10.5194/acp-12-7087-2012, 2012.
- Lee, S., Song, C. H., Park, R. S., Park, M. E., Han, K. M., Kim, J., Choi, M., Ghim, Y. S. and
 Woo, J.-H.: GIST-PM-Asia v1: development of a numerical system to improve particulate
 matter forecasts in South Korea using geostationary satellite-retrieved aerosol optical data
 over Northeast Asia, Geosci. Model Dev., 9(1), 17–39, doi:10.5194/gmd-9-17-2016, 2016.
- Levelt, P. F., Khattatov, B. V., Gille, J. C., Brasseur, G. P., Tie, X. X. and Waters, J. W.:
 Assimilation of MLS ozone measurements in the global three-dimensional chemistry
 transport model ROSE, Geophys. Res. Lett., 25(24), 4493–4496,
 doi:10.1029/1998GL900152, 1998.
- Lorenc, A. C.: Analysis methods for numerical weather prediction, Q.J.R. Meteorol. Soc.,
 112(474), 1177–1194, doi:10.1002/qj.49711247414, 1986.
- Louis, J.-F.: A parametric model of vertical eddy fluxes in the atmosphere, Boundary-Layer
 Meteorol, 17(2), 187–202, doi:10.1007/BF00117978, 1979.
- Malm, W. C. and Hand, J. L.: An examination of the physical and optical properties of aerosols
 collected in the IMPROVE program, Atmospheric Environment, 41(16), 3407–3427,
 doi:10.1016/j.atmosenv.2006.12.012, 2007.
- Martin, R. V., Jacob, D. J., Yantosca, R. M., Chin, M. and Ginoux, P.: Global and regional decreases in tropospheric oxidants from photochemical effects of aerosols, J. Geophys.
 Res., 108(D3), 4097, doi:10.1029/2002JD002622, 2003.
- NCL: The NCAR Command Language (Version 6.6.2) [Software]. Boulder, Colorado:
 UCAR/NCAR/CISL/TDD. http://dx.doi.org/10.5065/D6WD3XH5, 2019.
- Niu, G.-Y., Yang, Z.-L., Mitchell, Kenneth. E., Chen, F., Ek, M. B., Barlage, M., Kumar, A.,
 Manning, K., Niyogi, D., Rosero, E., Tewari, M. and Xia, Y.: The community Noah land
 surface model with multiparameterization options (Noah-MP): 1. Model description and
 evaluation with local-scale measurements, J. Geophys. Res., 116(D12109),
 doi:10.1029/2010JD015140, 2011.
- 799 Otte, T. L. and Pleim, J. E.: The Meteorology-Chemistry Interface Processor (MCIP) for the

- CMAQ modeling system: updates through MCIPv3.4.1, Geoscientific Model
 Development, 3(1), 243–256, doi:https://doi.org/10.5194/gmd-3-243-2010, 2010.
- Park, M. E., Song, C. H., Park, R. S., Lee, J., Kim, J., Lee, S., Woo, J.-H., Carmichael, G. R.,
 Eck, T. F., Holben, B. N., Lee, S.-S., Song, C. K. and Hong, Y. D.: New approach to
 monitor transboundary particulate pollution over Northeast Asia, Atmos. Chem. Phys.,
 14(2), 659–674, doi:10.5194/acp-14-659-2014, 2014.
- Park, R. S., Song, C. H., Han, K. M., Park, M. E., Lee, S.-S., Kim, S.-B. and Shimizu, A.: A
 study on the aerosol optical properties over East Asia using a combination of CMAQsimulated aerosol optical properties and remote-sensing data via a data assimilation
 technique, Atmos. Chem. Phys., 11(23), 12275–12296, doi:10.5194/acp-11-12275-2011,
 2011.
- Penner, J. E., Dong, X. and Chen, Y.: Observational evidence of a change in radiative forcing
 due to the indirect aerosol effect, Nature, 427(6971), 231–234, doi:10.1038/nature02234,
 2004.
- Pleim, J. E.: A Combined Local and Nonlocal Closure Model for the Atmospheric Boundary
 Layer. Part I: Model Description and Testing, J. Appl. Meteor. Climatol., 46(9), 1383–
 1395, doi:10.1175/JAM2539.1, 2007a.
- Pleim, J. E.: A Combined Local and Nonlocal Closure Model for the Atmospheric Boundary
 Layer. Part II: Application and Evaluation in a Mesoscale Meteorological Model, J. Appl.
 Meteor. Climatol., 46(9), 1396–1409, doi:10.1175/JAM2534.1, 2007b.
- 820
 Pleim, J. E. and Xiu, A.: Development of a Land Surface Model. Part II: Data Assimilation, J.

 821
 Appl.
 Meteor.,
 42(12),
 1811–1822,
 doi:10.1175/1520

 822
 0450(2003)042<1811:DOALSM>2.0.CO;2, 2003.
- Scott, C. E., Rap, A., Spracklen, D. V., Forster, P. M., Carslaw, K. S., Mann, G. W., Pringle, K.
 J., Kivekäs, N., Kulmala, M., Lihavainen, H. and Tunved, P.: The direct and indirect radiative effects of biogenic secondary organic aerosol, Atmos. Chem. Phys., 14(1), 447– 470, doi:10.5194/acp-14-447-2014, 2014.
- Skamarock, C., Klemp, B., Dudhia, J., Gill, O., Barker, D., Duda, G., Huang, X., Wang, W. and
 Powers, G.: A Description of the Advanced Research WRF Version 3, ,
 doi:10.5065/D68S4MVH, 2008.
- Tang, Y., Chai, T., Pan, L., Lee, P., Tong, D., Kim, H.-C. and Chen, W.: Using optimal interpolation to assimilate surface measurements and satellite AOD for ozone and PM2.5: A case study for July 2011, Journal of the Air & Waste Management Association, 65(10), 1206–1216, doi:10.1080/10962247.2015.1062439, 2015.
- Tang, Y., Pagowski, M., Chai, T., Pan, L., Lee, P., Baker, B., Kumar, R., Delle Monache, L.,
 Tong, D. and Kim, H.-C.: A case study of aerosol data assimilation with the Community
 Multi-scale Air Quality Model over the contiguous United States using 3D-Var and

- optimal interpolation methods, Geosci. Model Dev., 10(12), 4743–4758,
 doi:10.5194/gmd-10-4743-2017, 2017.
- Wiedinmyer, C., Quayle, B., Geron, C., Belote, A., McKenzie, D., Zhang, X., O'Neill, S. and
 Wynne, K. K.: Estimating emissions from fires in North America for air quality modeling,
 Atmospheric Environment, 40(19), 3419–3432, doi:10.1016/j.atmosenv.2006.02.010,
 2006.
- Wiedinmyer, C., Akagi, S., Yokelson, R., Emmons, L., Al-Saadi, J., Orlando, J. and Soja, A.:
 The Fire INventory from NCAR (FINN): A High Resolution Global Model to Estimate
 the Emissions from Open Burning, Geoscientific Model Development, 625–641, 2011.
- Woo, J.-H., Choi, K.-C., Kim, H. K., Baek, B. H., Jang, M., Eum, J.-H., Song, C. H., Ma, Y.I., Sunwoo, Y., Chang, L.-S. and Yoo, S. H.: Development of an anthropogenic emissions
 processing system for Asia using SMOKE, Atmospheric Environment, 58, 5–13,
 doi:10.1016/j.atmosenv.2011.10.042, 2012.
- Xiu, A. and Pleim, J. E.: Development of a Land Surface Model. Part I: Application in a
 Mesoscale Meteorological Model, J. Appl. Meteor., 40(2), 192–209, doi:10.1175/15200450(2001)040<0192:DOALSM>2.0.CO;2, 2001.
- Yang, Z.-L., Niu, G.-Y., Mitchell, Kenneth. E., Chen, F., Ek, M. B., Barlage, M., Longuevergne,
 L., Manning, K., Niyogi, D., Tewari, M. and Xia, Y.: The community Noah land surface
 model with multiparameterization options (Noah-MP): 2. Evaluation over global river
 basins, J. Geophys. Res., 116(D12110), doi:10.1029/2010JD015139, 2011.
- Yu, H., Dickinson, R. E., Chin, M., Kaufman, Y. J., Holben, B. N., Geogdzhayev, I. V. and
 Mishchenko, M. I.: Annual cycle of global distributions of aerosol optical depth from
 integration of MODIS retrievals and GOCART model simulations, J. Geophys. Res.,
 108(D3), 4128, doi:10.1029/2002JD002717, 2003.
- Yuan, H., Dai, Y., Xiao, Z., Ji, D. and Shangguan, W.: Reprocessing the MODIS Leaf Area
 Index products for land surface and climate modelling, Remote Sensing of Environment,
 115(5), 1171–1187, doi:10.1016/j.rse.2011.01.001, 2011.



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. NCL (2019) was used to draw this figure.

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874 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
- 876 CMAQ outputs with satellite-retrieved and ground-measured observations. The data process
- 877 for preparing the ICs is shown in the box with gray-dashed lines.



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 SO₂. The DA of NO₂ is excluded in the current study,

the reason for which is discussed in the text.



883 SULFATE NITRATE AMMONIUM OA EC OTHERS

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/m³ and 19.9 µg/m³, 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) SO₂, and (e) O₃ 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.



Figure 6. Aggregated average concentrations of (a) PM₁₀, (b) PM_{2.5}, (c) CO, and (d) SO₂ 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.



Figure 7. Comparison of CMAQ-simulated O₃ mixing ratios (BASE RUN with blue lines and
 DA RUN with red lines) with O₃ mixing ratios from Air Korea stations (open black circles).
 DA RUN was carried out by assimilating CMAQ outputs with Air Korea observations using

904 (a) only O₃ mixing ratios and (b) both O₃ and NO₂ 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.











Figure 10. Spatial distributions (first and second columns) and bias (third and fourth columns)
of (a) PM₁₀, (b) PM_{2.5}, (c) CO, (d) SO₂, and (e) O₃ over Seoul Metropolitan Area (SMA) for
the entire period of the KORUS-AQ campaign. Colored circles of first and second columns
represent the concentrations of the air pollutants observed at the Air Korea stations in the SMA.



Figure 11. Time-series plots of four performance metrics (IOA, R, RMSE, and MB) for (a) PM₁₀, (b) PM_{2.5}, (c) CO, (d) SO₂, and (e) O₃ forecasts. The DA was conducted at 00:00 UTC. The units of RMSE and MB are μ g/m³ and ppmv for PM concentrations and for gaseous species, respectively. The definitions of the four performance metrics are shown in Appendix A.





Figure 12. Variations of three performance metrics (R, RMSE, and MB) with time-intervals of 928 data assimilations. For these tests, the GOCI AODs were used in the DA to update the initial 929 conditions of the CMAQ model simulations. The results from the three CMAQ model 930 simulations were compared with AERONET AODs ("ground truth"). The two blue squares 931 represent the performances from the BASE RUNs and the red squares indicate the 932 performances from the DA RUNs. The three experiments were carried out with the assimilation 933 time-intervals of 24, 6, and 3 hours (hr), respectively. Here, the DA RUN with the 24-hr time-934 interval is referred to as "air quality prediction", and the DA RUNs with the 6-hr and 3-hr time-935 interval are referred to as "air quality reanalysis". 936



Figure 13. Soccer plot analyses for (a) PM₁₀, (b) PM_{2.5}, (c) CO, (d) SO₂, and (e) O₃. 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.

	PM ₁₀		PM _{2.5}		СО		SO ₂		O ₃	
	BASE RUN	DA RUN	BASE RUN	DA RUN	BASE RUN	DA RUN	BASE RUN	DA RUN	BASE RUN	DA RUN
Ν	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

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}		СО		SO ₂		0 ₃	
	BASE RUN	DA RUN	BASE RUN	DA RUN	BASE RUN	DA RUN	BASE RUN	DA RUN	BASE RUN	DA RUN
Ν	1057		695		1024		1007		1043	
ΙΟΑ	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

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