

Cover letter

12 November, 2019

Dr. Havala Pye
Topical editor of Geoscientific Model Development (GMD)
National Exposure Research Laboratory
U.S. Environmental Protection Agency
United States

Attn: Dr. Havala Pye (Topical editor)

Re: Manuscript (gmd-2019-169)

Dear Dr. Havala Pye:

Please find our revised manuscript titled “Development of Korean Air Quality Prediction System 1 version 1 (KAQPS v1) with focuses on practical issues” and our responses to both reviewers’ comments. First of all, we would like to express our thanks to both reviewers for their valuable suggestions and comments on our manuscript. Based on both reviewers’ suggestion and comments, we further improved the manuscript. The modified/added parts are painted in a red color in the revised manuscript.

Again, thank you for your consideration of this manuscript, and we look forward to hearing from you regarding the final decision of this paper

Sincerely,

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Response to reviewer 1

Authors appreciate reviewer's thoughtful comments and suggestions, which are greatly helpful for us to improve our manuscript. The manuscript has been revised to accommodate the reviewer's comments.

General comment:

This is a straightforward manuscript describing a WRF-CMAQ system with data assimilation adjustment for its initial condition, and shows that the data assimilation improve the predictions in most cases. Here are some specified comments.

Comments and response:

Comment: The data assimilation method mentioned mainly include AOD assimilation and surface measurement assimilation. Which one play a more important role for PM₁₀ and PM_{2.5} adjustment? Do these adjustments have any conflict?

Response: To clarify the impacts of the AOD and PM adjustments, assimilation was conducted in the following procedure. First, CMAQ simulated-AODs were assimilated with GOCI-retrieved AODs, and the assimilated AODs were then allocated into the PM concentrations, based on model-simulated concentrations. After that, the allocated PM was again assimilated using ground-based PM observations. Therefore, the assimilation using the surface measurements played a more important role in the entire PM adjustments. The reasons why the GOCI AODs were applied prior to using ground-based observations are two-fold: (1) GOCI slightly underestimates AODs compared to the AODs measured by the AERONET, which possibly leads to underestimated PM adjustments and (2) the allocation of AOD into PM concentrations based on model values has uncertainties. Despite these two reasons, we found that GOCI AODs are still useful for data assimilation (DA), because satellites provide meaningful information especially over the ocean areas where no surface-based observations are available.

Comment: Table 1 shows that CO's R and SO₂'s MNB become worse after data assimilation, why?

Response: As listed in Table 1 (in this response), the model-calculated CO concentrations (BASE RUN) are by far lower than those observed by in-situ measurements. After conducting DA at 00:00 UTC, the CMAQ-simulated CO concentrations became closer to observations. Up to 6 hours, the DA RUN showed a better performance (R=0.56; MB=0.017) compared to the BASE RUN (R=0.40; MB=-0.27). However, the differences between the BASE RUN and the DA RUN were diminished as the prediction progressed, because model tends to go back to its original state. Because of this tendency, the scatter plot of the DA RUN became more widespread, i.e., smaller correlation coefficient, than those of the BASE RUN for 0 – 23 hours predictions.

In case of SO₂ (see Table 2 in this response), the DA RUN showed a better performance for 0 – 6 hours and 0 – 23 hours predictions compared with the BASE RUN in terms of R, IOA, RMSE, and MB. Unlike these statistical variables, MNB, a relative difference normalized by observations, was decreased in the DA RUN for 0 – 6 hours predictions and increased for 0 – 23 hours predictions. Figure 11(d) shows the discrepancy of MB between daytime and nighttime. The model-simulated SO₂ concentrations of both the BASE RUN and the DA RUN were much smaller than observations during the daytime, and became similar (BASE RUN) or larger (DA RUN) compared to observations during the nighttime. These over-predicted nocturnal SO₂ concentrations of the DA RUN lead to large positive MNB values. This can also be explained by the underestimated nocturnal mixing layer height (MLH) shown in Fig. 8. For further investigation, we are collecting and analyzing more lidar data available over South Korea. In the future, a further comparison study will be carried out using those lidar-measured MLH over South Korea.

Table 1. Statistical metrics for CO from BASE RUN and DA RUN with Air Korea observations at 00:00 UTC when the DA was conducted, 0 – 6 hr predictions after DA, and 0 – 23 hr predictions over the entire period of the KORUS-AQ campaign.

CO	At DA time (00 UTC)		0 - 6 hr prediction		0 - 23 hr prediction	
	BASE RUN	DA RUN	BASE RUN	DA RUN	BASE RUN	DA RUN
N	1024		27268		101764	
IOA	0.41	0.62	0.24	0.33	0.41	0.51
R	0.28	0.43	0.40	0.56	0.28	0.21
RMSE	0.35	0.16	0.31	0.17	0.31	0.19
MB	-0.31	-0.01	-0.27	0.017	-0.27	-0.04
MNB	-64.3	9.69	-62.52	17.11	-62.0	3.14

Table 2. Same as Table 1, except for SO₂.

SO ₂	At DA time (00 UTC)		0 - 6 hr prediction		0 - 23 hr prediction	
	BASE RUN	DA RUN	BASE RUN	DA RUN	BASE RUN	DA RUN
N	1007		27258		101764	
IOA	0.36	0.44	0.34	0.37	0.34	0.35
R	0.097	0.27	0.13	0.15	0.14	0.15
RMSE	0.0061	0.0039	0.0074	0.0065	0.0068	0.0066
MB	-0.0019	-0.0009	-0.0021	-0.0014	-0.0009	-0.0004
MNB	-20.1	7.35	-29.87	-7.54	3.1	17.77

Comment: Line 276 (page 12), “Tang et al., 2017” cannot be found in the reference.

Response: “Tang et al., 2017” has been included to the references. Please, see pp. 34:834 – pp. 35: 838.

Comment: Line 282 (page 13), equation (7): the term “I” does not come with explanation in the text.

Response: The sentence of “I denotes the unit matrix” has been added to the manuscript. Please, see pp. 14:320 – pp. 15:321.

Response to reviewer 2

Authors appreciate reviewer's thoughtful comments and suggestions, which are greatly helpful for us to improve our manuscript. The manuscript has been revised to accommodate the reviewer's comments.

General comment and response:

General Comments: This paper discussed an application of offline WRF-CMAQ simulations and evaluation both with and without DA during the KORUS-AQ measurement campaign in Korea during May 1 – June 12, 2016. While the paper is well written, and the DA methodology is sound, there are some concerns on missing details and discussion throughout the paper (see Specific comments), rather expected model performance results, and using a such a case study/campaign application as an appropriate demonstration for the readiness of an “operational” air quality forecast system (below). While I recommend accepting the article for publication, these issues should be addressed.

I think it is a bit misleading to call this application of WRF-CMAQ simulations a true “operational” air quality forecasting system, as it is currently only applied for a very short time period (May 1 – June 12) during a detailed measurement campaign. Such detailed observations are likely not available to demonstrate a continuous refresh of satellite/surface observations for DA in an operational system. To support this model system's use as “operational”, what confidence is there that the statistical performance will scale to the remainder of the calendar year under different meteorological and chemical (i.e., emissions) environments? I think the “operational” focus of the paper should be dialed back in the paper, in place of a focus on the application of offline WRF-CMAQ chemical DA and evaluation during an intensive measurement campaign in Korea. Towards the end of the paper, it could be discussed of this WRF-CMAQ configuration could be further developed and more comprehensively tested to become an operational air quality forecasting system for Korea.

This issue is compounded by the somewhat expected results in the paper of improved

model performance by assimilating data (compared to no DA), the impacts of a higher frequency rate of DA, and lack of testing the important “trade-off” that exists between increased precision and computational cost of DA. In fact, the authors explicitly state that this unsurprising result needs to be further tested, and that the true operational system should be designed under these considerations. These facts further substantiate my concern that it may be premature to consider this paper a description of an “operationally ready” model, but rather an application of testing DA in CMAQ during a relatively intensive KORUS campaign, which is a region that would certainly benefit from further development and testing of an operational air quality forecast model.

Response: Since we agreed that further researches are needed for true “operational” air quality prediction system, the word of “operational” has been removed from the original manuscript, and the title of the paper has also been modified to “Development of Korean Air Quality Prediction System version 1 (KAQPS v1) with focuses on practical issues” in order to avoid any confusion.

Actually, based on this work, the multiple-year tests are being currently conducted with the current sets of WRF-CMAQ-DA system. Once these tests are finished, we will revisit and report the issue of the “operational” air quality prediction system in South Korea, again.

Specific comments and response:

Comment: S1. Lines 88 – 93: Don’t like the interchanging of air quality and chemical weather terminology here, as it is too similar to suggest that online chemical weather feedbacks are necessary in air quality models. I suggest revising to avoid any confusion.

Response: All of the terminology “chemical weather” has been replaced with “air quality” in the manuscript to avoid this confusion. Please, check out pp. 4:85 – 90.

Comment: S2. Lines 95 – 96: This is partially being overcome by new high spatial and temporal resolution satellite observations of composition (e.g., TEMPO, TROPOMI, GEMS, etc.). I think although they lack the longer term records, it should be mentioned that strides are being made at tackling these issues for air composition observations.

Response: We totally agree with referee's comment. A paragraph has been added to the revised paper for explaining the efforts to improve spatial and temporal coverage of satellite measurements. Please, refer to the added parts (pp. 5:93 – 101).

Comment: S3. Lines 98-101: What about the issues of coarse model grid scale for CTMs?

Response: Coarse-grid scale for CTM simulations can increase model uncertainties for several cases (Shrestha et al., 2009; Sirithian and Thepanondh, 2016), but not for all. After consideration of the matter, we have decided not to address this issue in detail, because it is beyond the scope of our manuscript.

Comment: S4. Lines 142-155: This section is lacking information.

1) What are the dynamical/physical configurations for WRF (e.g., LSM, land use data, sfc layer, PBL, grid scale microphysics, convective cloud parameterization, etc.) ?

Response: The dynamical and physical configurations for the WRF model simulations were added in the revised paper. Please, see pp. 7:151 – 157.

2) The met processor needs to be defined (i.e., MCIP) and explained for important derived variables from WRF to drive CMAQ. Many physical inconsistencies can arise between WRF-MCIPCMAQ, and this is pivotal information in understanding the physical linkages between the upstream physics in WRF to drive CMAQ.

Response: Information of MCIP has been added to the manuscript. Please, check out pp. 8:169 – 174.

3) How exactly is OBSGRID applied in this model, and how does it relate to the WRF physical configurations?

Response: The OBSGRID does not relate to the WRF physical configurations. This process was conducted to improve the accuracy of initial and boundary conditions for the WRF model simulation via data assimilation technique. A short sentence explaining OBSGRID has been added with more detailed information. Please, check this out at pp. 8:160 – 165.

4) Why was 15x15 km chosen, as opposed to commonly applied forecast models at 12x12 km? While much of this could be provided in supplemental/appendix to preserve brevity in the main text, it still needs to be included somewhere in the manuscript.

Response: Based on Lee et al. (2016)'s work, we chose 15 km by 15 km for CMAQ.

Comment: S5. Lines 157-177: This section is also lacking information.

1) What are the main chemical configurations for CMAQ (e.g., gas-phase chemistry, aerosol mechanism/size, dust/sea-salt, aqueous phase, dry/wet deposition, etc.)?

Response: The main chemical and physical configurations for the CMAQ model simulations were added into the revised manuscript. Please, refer to pp. 8:180 – pp. 9:189.

2) How does the CMAQ configurations interact and physically link with the upstream, driving physical configurations of WRF (comment S4)?

Response: Information on physical configurations of WRF and CMAQ model simulations has been added to the revised manuscript (refer to the responses to comments S4 and S5).

3) Clarification is needed on why MEGAN, rather than in-line BEIS, was used for biogenics in CMAQ. Is this based on literature for the Korean region?

Response: MEGAN was applied to the CMAQ model simulations in this study, because the MEGAN has been utilized in Korean modeling community and has also been widely used in many studies focusing on East Asia including South Korea (Kim et al., 2014; Kim et al., 2017; Lee et al., 2016; Park et al., 2014; Sourì et al., 2017).

Comment: S6. Lines 230 – 255: I think this is an important discussion, but please make it clear how much this is a different formulation of AOD compared to the other pre-existing AOD calculations in CMAQv5.1 (e.g., the reconstruction method).

Response: A paragraph has been added in the revised paper. Please, see pp. 13:277 – 286.

Comment: S7. Lines 311 – 316: I have some issue with assuming that $\Delta PM_{2.5}$ exactly scales with ΔPM_{10} , because the inherent differences in some of the sources that make up the PM_{10} bias compared to $PM_{2.5}$. In other words, if most of the ΔPM_{10} is due to missing coarse mode aerosol emissions (e.g., dust etc.), we wouldn't expect this difference to have the same effect on $\Delta PM_{2.5}$.

Response: Yes, it may be a correct point! Our problem has been that the missing sources of the coarse-mode discrepancy (i.e., $\Delta PM_{2.5-10}$) have not been identified in South Korea. We are thinking that the uncertainty in the fugitive dust emissions from construction sites, road sites, cattle-raising areas, dry mud fields, etc may take significant responsibility for the $\Delta PM_{2.5-10}$ in South Korea. However, the amounts of such emissions from individual source have been difficult to quantify. In addition to a method we used in this study, we have to add the above amounts in the future study. This may be the reason why Fig. 9 shows larger differences in the PM_{10} predictions than in the $PM_{2.5}$ predictions.

Comment: S8. Lines 391 – 392: Could this also be due to underpredicted NO₂ with the DA run and not enough nighttime ozone titration? This perhaps could be better explored using Ox relationships and looking into different regions of the domain.

Response: A following sentence has been added into the revised manuscript. Please, see pp. 19:424 – 428.

Comment: S9. Lines 397 – 398: It is concerning to draw such a conclusion based only on a single SNU lidar site comparison. Is this truly a widespread issue for nocturnal boundary layers in Korea? While this may indeed be common and well-defined previously using similar WRF physical options as in this study, there needs to be appropriate references here to provide support for your argument.

Response: Although we showed only one site example here, this nocturnal MLH problem has been commonly found in South Korea. Korean modeling community have also been well aware of this problem for a long time (Kang et al., 2016; Nam et al., 2016). A sentence in Lines 397-398 of the original manuscript has been modified, because a more intensive comparison study between lidar-retrieved and model-simulated MLH is necessary. Please, see pp. 20:435 – 437.

Comment: S10. Lines 406 – 408: This methodology is not clear. What set of MLH observations would be used for this effort? It certainly cannot be based on the single SNU lidar site. How would this be done “operationally” in the offline MCIP step between WRF and CMAQ? Bias correcting the MLH may lead to physical inconsistencies with other driving meteorological fields from WRF that were based on a particular set of physical configurations. Overall, this requires more thought likely, raises some concern, and should probably just be removed from the paper.

Response: Following reviewer’s suggestion, the related sentences of Lines 406-408 have been removed from the manuscript since we agree that careful consideration of

the MLH bias correction is needed. We are thinking that the bias correction will not lead to physical inconsistency in the “off-line” mode modeling, but it can create a problem in the “on-line (two-way)” modeling.

Comment: S11. Lines 412 - 413: CMAQ already has the “capability” to predict aerosol composition. Thus, it should be restated to say a “: :a strong capability of our DA system is to improve predictions of CMAQ aerosol composition”.

Response: We modified the corresponding sentence into “a strong capability of our DA system is to improve predictions of CMAQ aerosol composition”. Please, check this out at pp. 20:441 – 442.

Comment: S12. Lines 416 – 424: These changes in model performance would be elucidated if an addition column showing the absolute bias difference plot (colored in Red Blue shading) for the two runs compared to surface observations in Figure 10. This can be achieved by interpolating the closest model to the observations points.

Response: Yes, it is a good idea! Following reviewer’s suggestion, bias difference plot has been added into Fig. 10, and the caption of the figure has also been changed. Please, check out the modified Fig. 10 at pp. 45 in the revised manuscript.

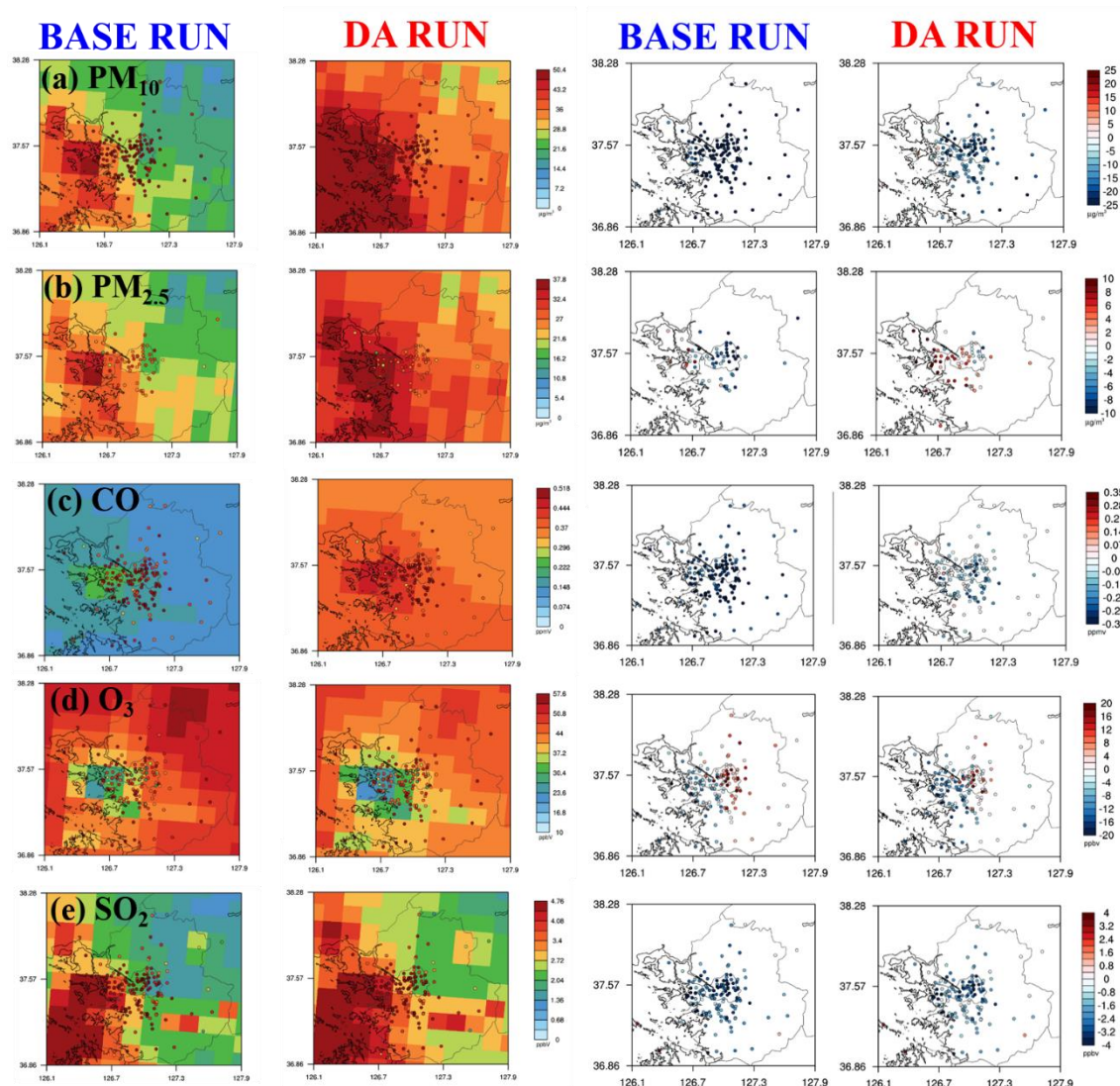


Figure 10. Spatial distributions (first and second columns) and bias (third and fourth columns) of (a) PM_{10} , (b) $\text{PM}_{2.5}$, (c) CO , (d) SO_2 , and (e) O_3 over Seoul Metropolitan Area (SMA) for the entire period of the KORUS-AQ campaign. Colored circles of first and second columns represent the concentrations of the air pollutants observed at the Air Korea stations in the SMA.

Comment: S13. Lines 428 – 429: This is confusing, as it appears you are talking about an additional model simulation to the DA run. I thought that the adjusted NO_x observations are used in the DA run, as discussed for the results in Lines 363-373 and comparing results from Figures 7a-b.

Response: As addressed in Lines 428-429 of the original manuscript, Air Korea observations for NO₂ were not applied in a 100 % accurate way in the current version of Korean air quality prediction system. It is because NO₂ mixing ratios measured at the Air Korea sites are contaminated by other nitrogen gases due to “NO₂ measurement artifacts” as discussed in Lines 374-387 (original manuscript). In Fig. 7(a), the results of a DA RUN by assimilating CMAQ outputs with Air Korea-observed O₃ mixing ratios are shown. Fig. 7(b) depicts the results of the test run by assimilating CMAQ outputs with Air Korea-observed both O₃ and NO₂ mixing ratios. But, here we used “0.8×NO₂ mixing ratios”, following information given by Jung et al. (2017). We believe this work is not 100 % perfect! That’s why we call the DA RUN a preliminary DA RUN. In the future, we attempt to correct these artifacts of NO₂ mixing ratios. Then, we will revisit the impacts of the NO₂ assimilation on O₃ mixing ratios. To avoid confusion regarding this issue, the corresponding sentence was modified in the revised manuscript (please, see pp. 18:394 – 396).

Comment: S14. Lines 525 – 529: Is the AI referring to its application to rapid refresh of emissions, chemical reaction/mechanism replacements, or something else? Is there a large body of research that shows AI can even "improve" air quality forecasts? I think the body of work shows that AI can be used speed up the gas chemistry in regional CTMs, while not suffering model performance degradation. Also, if the provided citation to Kim et al. (2019) is of no help, because it is not included in the reference list (see T6 correction below).

Response: Kim et al. (2019) recently published a paper in ACP that employed a deep recurrent neural network system based on long short-term memory (LSTM) model for daily PM₁₀ and PM_{2.5} predictions in South Korea. The prediction system was optimized by iterative model trainings with the inputs of ground-based observations for PM₁₀, PM_{2.5}, and the observed meteorological variables including wind speed, wind direction, relative humidity, precipitation, etc. Their AI-based prediction system showed better performances than the CMAQ model simulations. However, the current AI system works only for points where ground-based observations are made. Therefore, we

expect that a combination of the AI system with the currently developed air quality prediction system can produce a more accurate air quality forecast over South Korea. Regarding this issue, please refer to pp. 25:550 – 559.

Technical corrections and response:

Comment: T1. Line 120: Tang et al. (2017) is not found in the reference list.

Response: “Tang et al., 2017” has been added into the references. Please, check this out at pp. 34:834 – pp. 35: 838.

Comment: T2. Line 145: Replace “dynamic” with “dynamical”.

Response: “dynamic” has been replaced by “dynamical”. Please, check this out at pp. 7:149.

Comment: T3. Line 147: Replace “National Centers for Environmental Prediction Final Analysis data (NCEP FNL)” with “National Centers for Environmental Prediction (NCEP) Final (FNL) Operational Global Analysis data on $1^{\circ} \times 1^{\circ}$ grids”. Is $1^{\circ} \times 1^{\circ}$ correct?

Response: Yes, $1^{\circ} \times 1^{\circ}$ is correct. “National Centers for Environmental Prediction Final Analysis data (NCEP FNL)” has been replaced with “National Centers for Environmental Prediction (NCEP) Final (FNL) Operational Global Analysis data on $1^{\circ} \times 1^{\circ}$ grids”. Please, see this out pp. 7:157 – pp. 8:159.

Comment: T4. Line 413. Replace “matters” with “matter”

Response: “matters” has been deleted in the manuscript, following the referee’s advice (S11) . Thank you!

Comment: T5. Line 501: Replace “ground” with “near-surface”

Response: “ground” has been replaced with “near-surface”. Please, check this out at pp. 24:526 – 527.

Comment: T6. Line 527: Kim et al. (2019) is not found in the reference list.

Response: “Kim et al. (2019)” has been added to the references. Please, check this out at pp. 32:754 – 757.

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

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Short title: Air quality prediction system in Korea

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Abstract

For the purpose of providing reliable and robust air quality predictions, an air quality prediction system was developed for the main air quality criteria species in South Korea (PM_{10} , $\text{PM}_{2.5}$, 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 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 $\text{PM}_{2.5}$ (IOA = 0.71; MB = -2.43) as compared to the BASE RUN for PM_{10} (IOA = 0.51; MB = -27.18) and $\text{PM}_{2.5}$ (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 air

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 the measurement artifacts (i.e., inclusion of alkyl nitrates, HNO₃, and PANs in the ground-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 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 air quality prediction system (KAQPS).

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

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 (O₃), nitrogen dioxide (NO₂), 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 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 O₃ are the most notorious health threats, as has been stated by several previous studies (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 (PM₁₀, PM_{2.5}, O₃, CO, SO₂, and NO₂) 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 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 air quality forecasts are 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. These limitations will be overcome by improving spatial and temporal coverage via future geostationary satellite instruments such as the Geostationary Environment Monitoring Spectrometer (GEMS) over Asia, the Tropospheric Emissions: Monitoring of Pollution (TEMPO) over North America, and the Sentinel-4 over Europe. In addition, the Tropospheric 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 information on the chemical composition in the atmosphere with a higher spatial resolution of $3.5 \times 7 \text{ km}^2$.

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 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 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.

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.

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) dynamical core was applied to prepare the meteorological inputs for the CMAQ model simulations. Dynamical and physical configurations for the WRF model simulations were selected as follows: the Yonsei University (YSU) scheme for planetary boundary layer (Hong et al., 2006); 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 Noah-MP land surface model (Niu et al., 2011; Yang et al., 2011); the rapid radiative transfer 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 National Centers for Environmental Prediction (NCEP) Final (FNL) Operational Global

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.

The model domain for the WRF simulations covers Northeast Asia with a horizontal resolution of $15 \times 15 \text{ km}^2$, 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 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 transformed into the CMAQ-ready format via the Meteorology-Chemistry Interface Processor (MCIP; Otte and Pleim (2010)) v4.3 which is a software to serve for transforming horizontal and vertical coordinates while trying to maintain dynamic consistency between WRF and CMAQ model simulations.

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 $15 \times 15 \text{ km}^2$ 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 (Appel et al., 2013); Euler Backward Iterative (EBI) chemistry solver (Hertel et al., 1993), which is a numerically optimized photochemistry mechanism solver; M3DRY for dry 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); MULTISCALE (Louis, 1979), which is a simple first-order eddy diffusion scheme for horizontal diffusion; and the Asymmetric Convective Model 2 (ACM2; Pleim, 2007a, 2007b) for vertical diffusion.

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 $2500 \times 2500 \text{ km}^2$ (refer to Fig. 1). Among the three payloads of the COMS, Geostationary Ocean Color Image (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 $500 \times 500 \text{ m}^2$ 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 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 quality-assured level 2.0 AODs for the AERONET were used in this study.

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 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}) using the following equation:

$$\text{AOD}(\lambda) = \int_0^z \sigma_{\text{ext}}(\lambda) \, dz \quad (1)$$

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:

$$\sigma_{\text{ext}}(\lambda) = \sigma_{\text{abs}}(\lambda) + \sigma_{\text{sca}}(\lambda) \quad (2)$$

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

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

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 ; $[C]_{ij}$ is the concentration of particulate species including $(\text{NH}_4)_2\text{SO}_4$, NH_4NO_3 , black carbon, organic aerosols (OA), mineral dust, and sea-

salt aerosols; RH is the relative humidity; $f_{ij}(\text{RH})$ is the hygroscopic factor; and the single scattering albedo (ω) implies to the fraction (portion) of scattering in the total extinction.

Using Eqns. (2) - (4), AODs were calculated from the aerosol composition and RH. These have been intensive tests using different β and $f(\text{RH})$ values in the following three previous studies: (1) Chin et al. (2002)'s study with the Goddard Chemistry Aerosol Radiation and Transport (GOCART) model; (2) Martin et al. (2003)'s study with the GEOS-Chem model; and (3) Malm and Hand (2007)'s study with the CMAQ model. Lee et al. (2016) tested these 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 estimated with the β and $f(\text{RH})$ values suggested by Chin et al. (2002). After that, σ_{ext} was integrated with respect to altitude, in order to calculate the AODs. The calculated AODs were used in the air quality prediction system in order to prepare the 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 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, O₃, and SO₂ were combined with the concentrations of CO, O₃, and SO₂ 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 PM₁₀; (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 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) as follows:

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

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

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

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

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; \mathbf{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 (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., 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 PM₁₀ & PM_{2.5} into particulate composition

In the procedure of DA, PM₁₀ was assimilated in this study, because the PM₁₀ data were more plentiful than PM_{2.5}. The assimilated PM₁₀ 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, 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

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 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 PM₁₀, PM_{2.5}, CO, and SO₂ against the Air Korea observations. Our air quality prediction system re-generated relatively well-matched concentrations for PM₁₀, PM_{2.5}, and CO from the DA RUN.

Figure 7 shows the case of ozone from the DA RUN by assimilating CMAQ outputs with Air Korea-observed (a) O₃ mixing ratios, and (b) both O₃ and NO₂ mixing ratios for a 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 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 NO₂ 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 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 NO₂ mixing ratios.

Although the DA for NO₂ provided better ozone predictions, one should take caution in using the NO₂ observations. The NO₂ mixing ratios measured at Air Korea sites are known to be contaminated by other nitrogen gases such as nitric acid (HNO₃), peroxyacetyl nitrates (PANs), and alkyl nitrates (ANs), since the Air Korea NO₂ mixing ratios are measured through a chemiluminescent method with catalysts of gold or molybdenum oxide at high temperatures.

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 the Air Korea NO₂ data, and are then used to prepare the ICs via the DA for more accurate 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 of the NO₂ measurement artifacts, more evolved schemes of ozone and NO₂ predictions will be possible in the future. As shown in Fig. 7, about a 20% reduction (average fraction of non-NO₂ mixing ratios in the observed NO₂ mixing ratios) was made for these demonstration runs (Jung et al., 2017).

Another practical issue is now discussed. Although the assimilation with the observed 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 aggregated plot of the ozone mixing ratios at the observation sites (see Fig. 7). This can be caused by underestimated NO₂ mixing ratios and thus not enough nighttime ozone titration. As aforementioned, reliable NO₂ prediction via the correction of the NO₂ measurement artifacts 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 a comparison between lidar-measured MLH (black dashed line) and WRF-calculated MLH (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 times higher than the nocturnal WRF-calculated MLH as measured at a lidar site inside the campus of Seoul National University (SNU) in Seoul. 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. Based on this discrepancy shown in Fig. 8, more intensive comparison study is being carried out by comparing lidar-measured 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.

3.1.2. Spatial distribution

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 (SMA). Noticeable improvements are observed to have been achieved in the spatial distributions by applying the ICs into the CMAQ model simulations, particularly for PM₁₀ (Fig. 10a), PM_{2.5} (Fig. 10b), and CO (Fig. 10c). As shown in Fig. 10, the under-predicted concentrations of PM₁₀, PM_{2.5}, 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 observations than the BASE RUN, but there were still under-predicted SO₂ 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 O₃, 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 NO₂ 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 NO₂ 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 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 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 PM₁₀ (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 PM₁₀ (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 O₃ and SO₂, 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 PM₁₀ (R: 0.3→0.75; slope: 0.17→0.66) and O₃ (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

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) PM₁₀, (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

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 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, the air quality prediction system was developed by preparing the ICs for CMAQ model simulations using GOCI AODs and ground-based observations of PM₁₀, 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.

The performances of the developed prediction system were evaluated using near-surface 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 SO₂), 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 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 NO₂ mixing ratios and the corrections of model-simulated mixing layer height (MLH). For SO₂, the performances of both the BASE RUN and the DA RUN were somewhat poor. Regarding this issue, more accurate SO₂ emissions are required to achieve better SO₂ 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 $7 \times 8 \text{ km}^2$ 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 (AI)-based air quality prediction systems are also currently being developed in several ways (e.g., Kim et al., 2019). Actually, Kim et al. (2019) developed an AI-based PM prediction 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-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 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 both the CTM-based air quality prediction and the AI-based prediction systems, both systems 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.

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). NCL (2019; doi:10.5065/D6WD3XH5) was used to draw the figures.

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 air quality prediction system are defined as follows.

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

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

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

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

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

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

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

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

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

612

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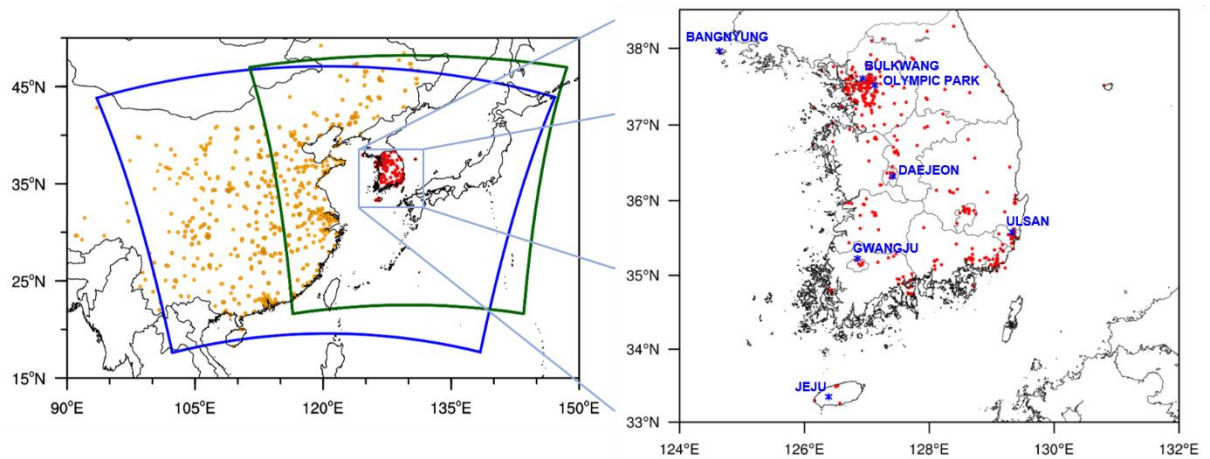


Figure 1. Domains of GOCI sensor (dark green line) and CMAQ model simulations (blue line). Red-colored dots denote the locations of Air Korea sites in South Korea. Orange-colored dots represent the locations of ground-based observation stations in China. Blue stars show the locations of seven super-sites 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. NCL (2019) was used to draw this figure.

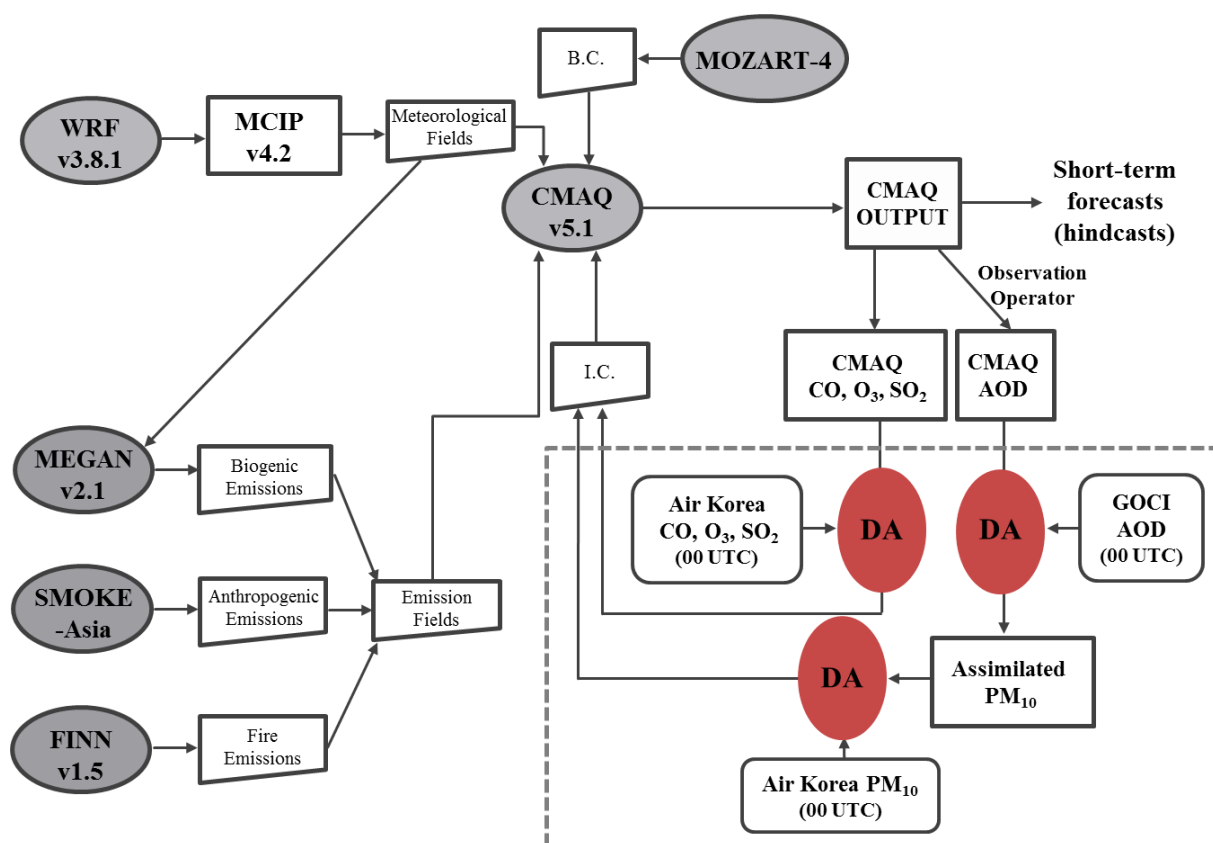


Figure 2. Schematic diagram of the Korean air quality prediction system developed in this study. The initial conditions (ICs) of the CMAQ model simulations are prepared by assimilating CMAQ outputs with satellite-retrieved and ground-measured observations. The data process for preparing the ICs is shown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

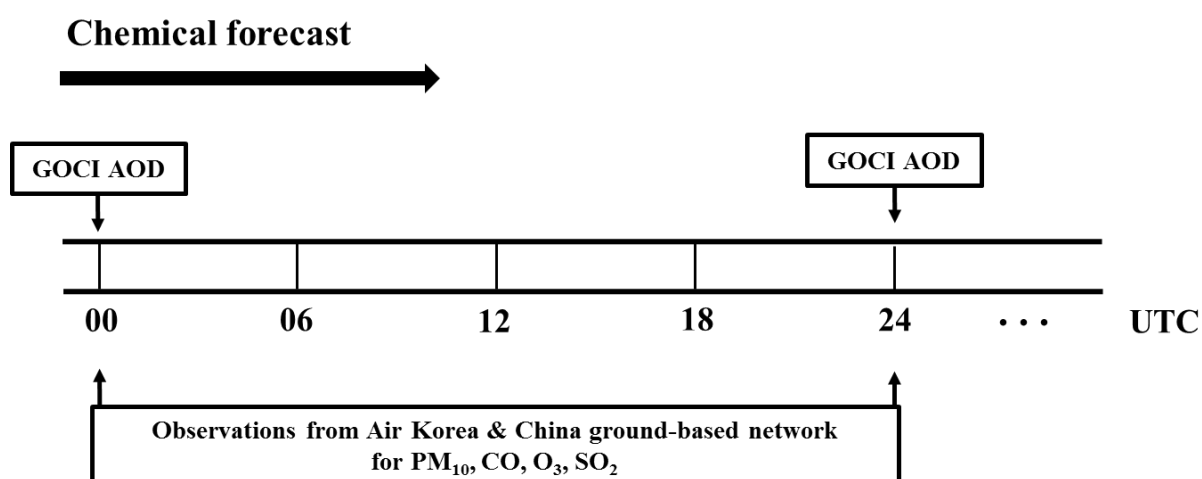
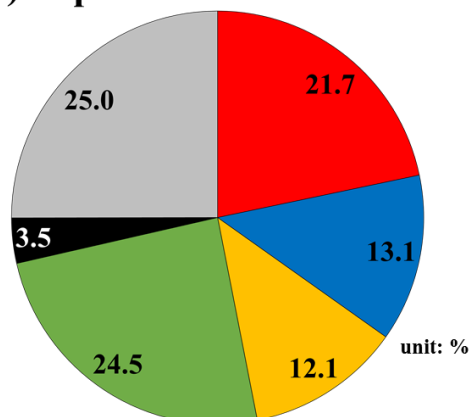


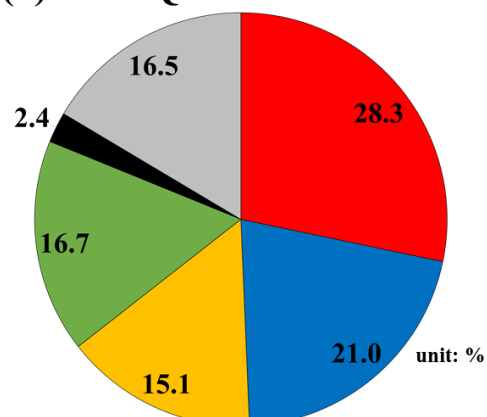
Figure 3. Schematic diagram of the Korean air quality prediction system for particulate matter (PM) and gas-phase pollutants. The data assimilation (DA) cycle is 24 hours for both PM and gas-phase pollutants such as CO, O₃, and SO₂. The DA of NO₂ is excluded in the current study, the reason for which is discussed in the text.

(a) Super-site



Average PM_{2.5} = 28 µg/m³

(b) CMAQ



Average PM_{2.5} = 19.9 µg/m³

■ 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.

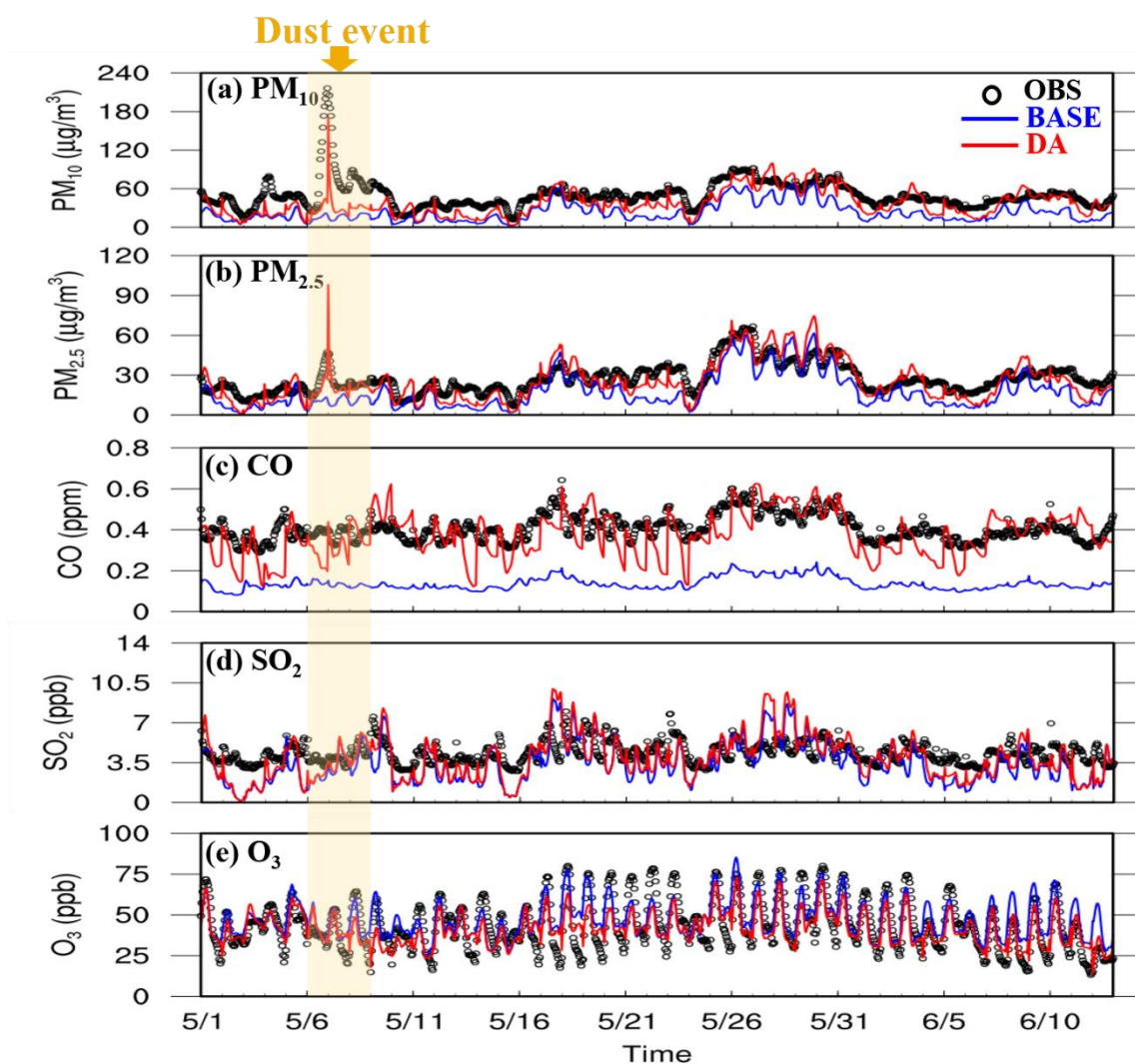


Figure 5. Time-series plots of hourly (a) PM_{10} , (b) $\text{PM}_{2.5}$, (c) CO, (d) SO_2 , and (e) O_3 concentrations at 264 Air Korea stations. Black open circles (OBS) represent the observed concentrations. Blue and red lines show the results simulated from the BASE RUN and DA RUN over South Korea,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

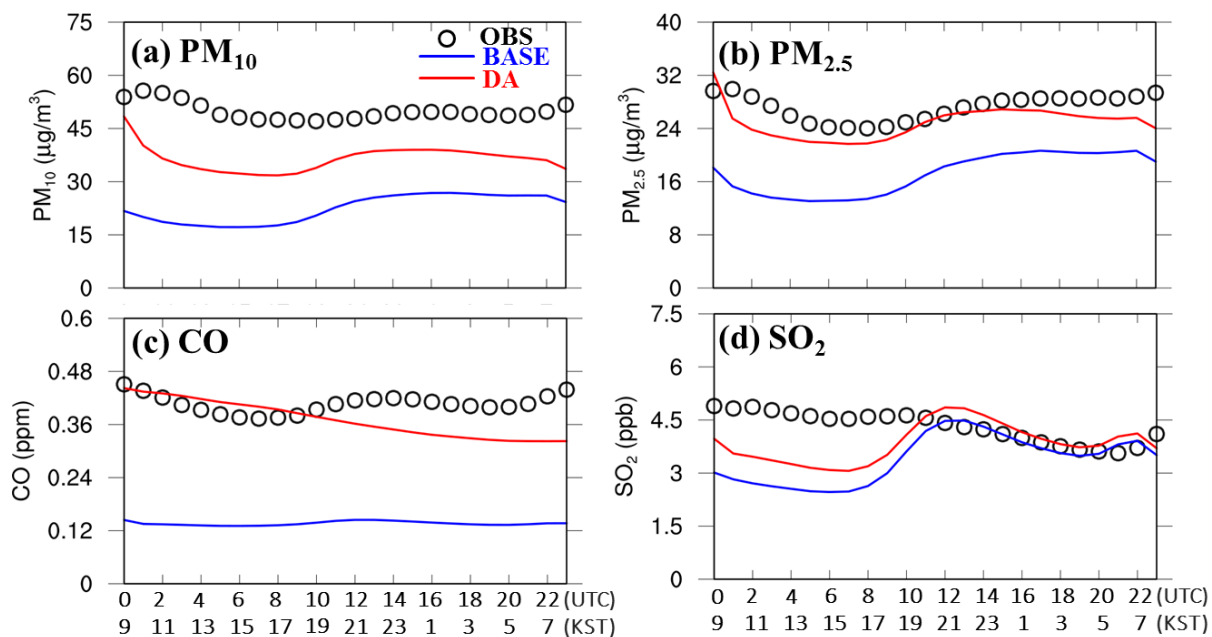


Figure 6. Aggregated average concentrations of (a) PM_{10} , (b) $\text{PM}_{2.5}$, (c) CO , and (d) SO_2 at 264 Air Korea stations over the KORUS-AQ campaign period. Open black circles denote the observations obtained from 264 Air Korea stations in South Korea. Blue and red lines represent the predicted 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.

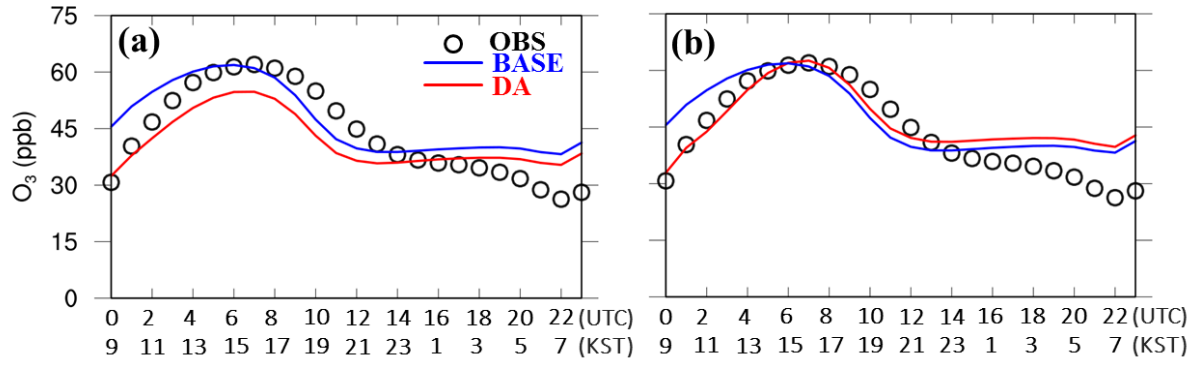


Figure 7. Comparison of CMAQ-simulated O₃ mixing ratios (BASE RUN with blue lines and DA RUN with red lines) with O₃ mixing ratios from Air Korea stations (open black circles). DA RUN was carried out by assimilating CMAQ outputs with Air Korea observations using (a) only O₃ mixing ratios and (b) both O₃ and NO₂ mixing ratios.

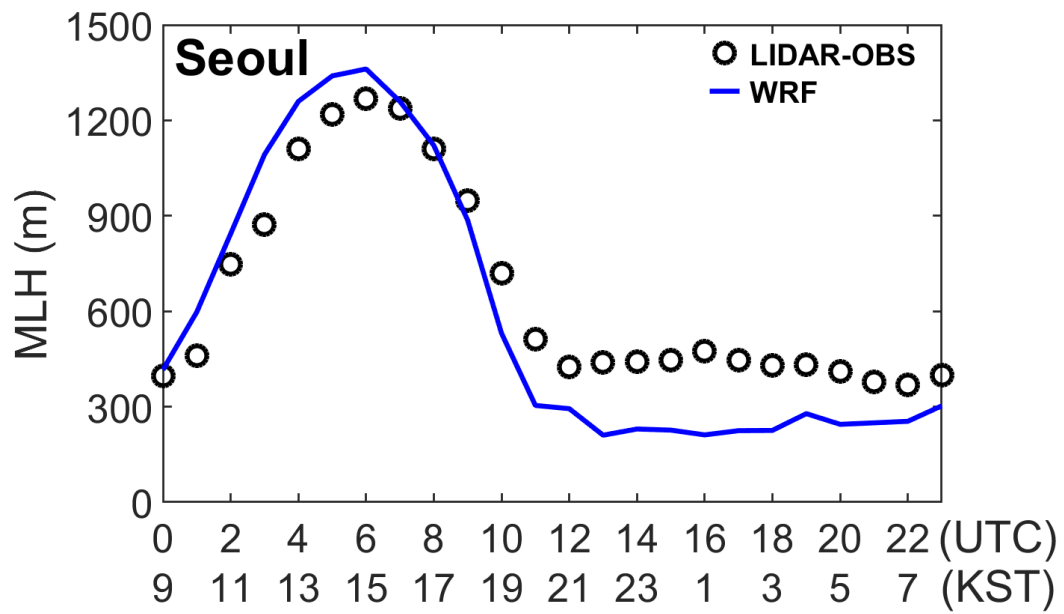


Figure 8. Comparison of WRF-simulated mixing layer height (MLH) (denoted by blue-dashed line) with lidar-measured MLH (denoted by open black circles) at Seoul National University (SNU) in Seoul. KST stands for Korean standard time.

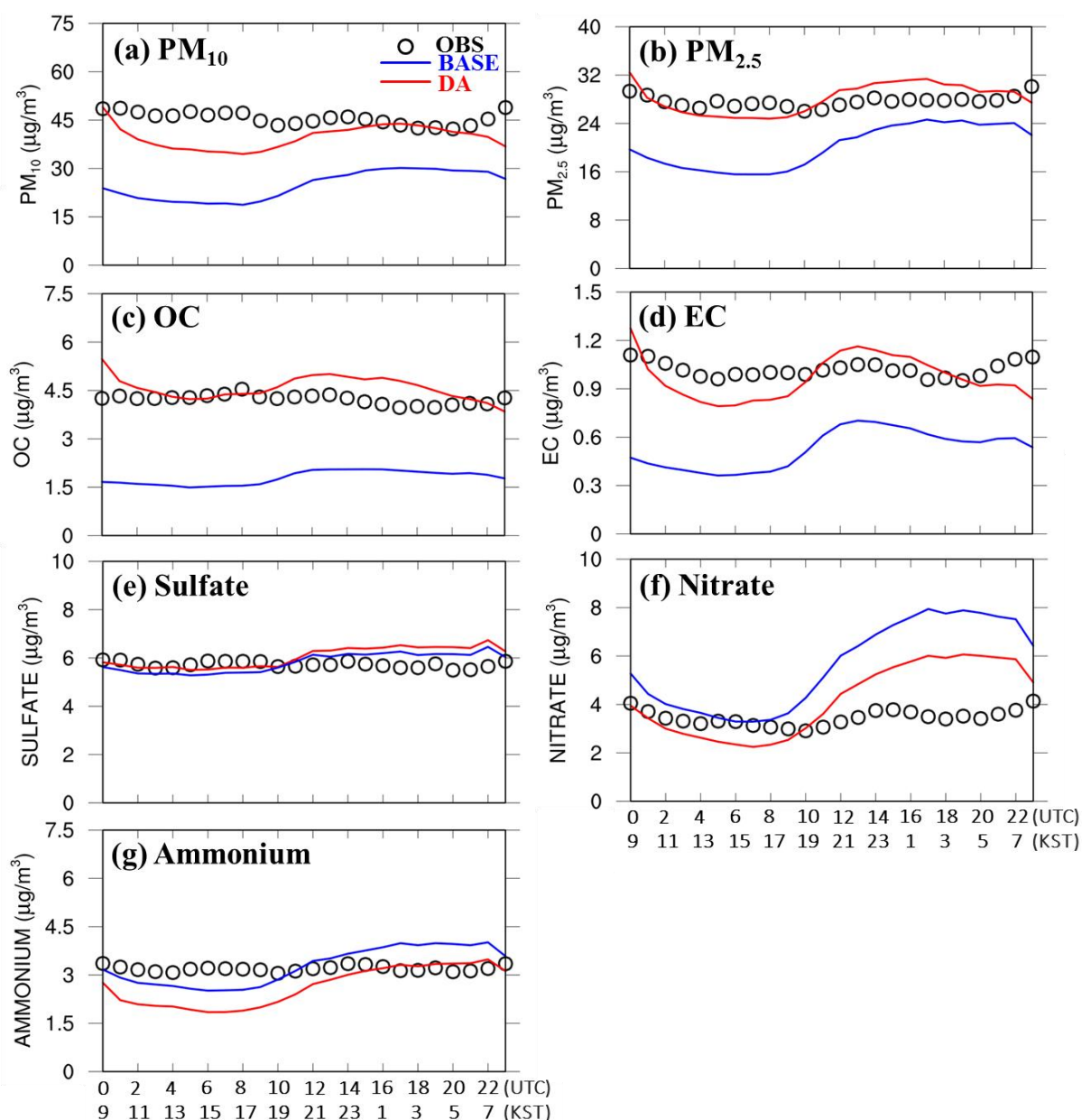


Figure 9. Aggregated average concentrations of (a) PM_{10} , (b) $\text{PM}_{2.5}$, (c) OC, (d) EC, (e) sulfate, (f) nitrate, and (g) ammonium as predicted by CMAQ model during the period of the KORUS-AQ campaign. The others are the same as those shown in Fig. 7, except for the fact that the observation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

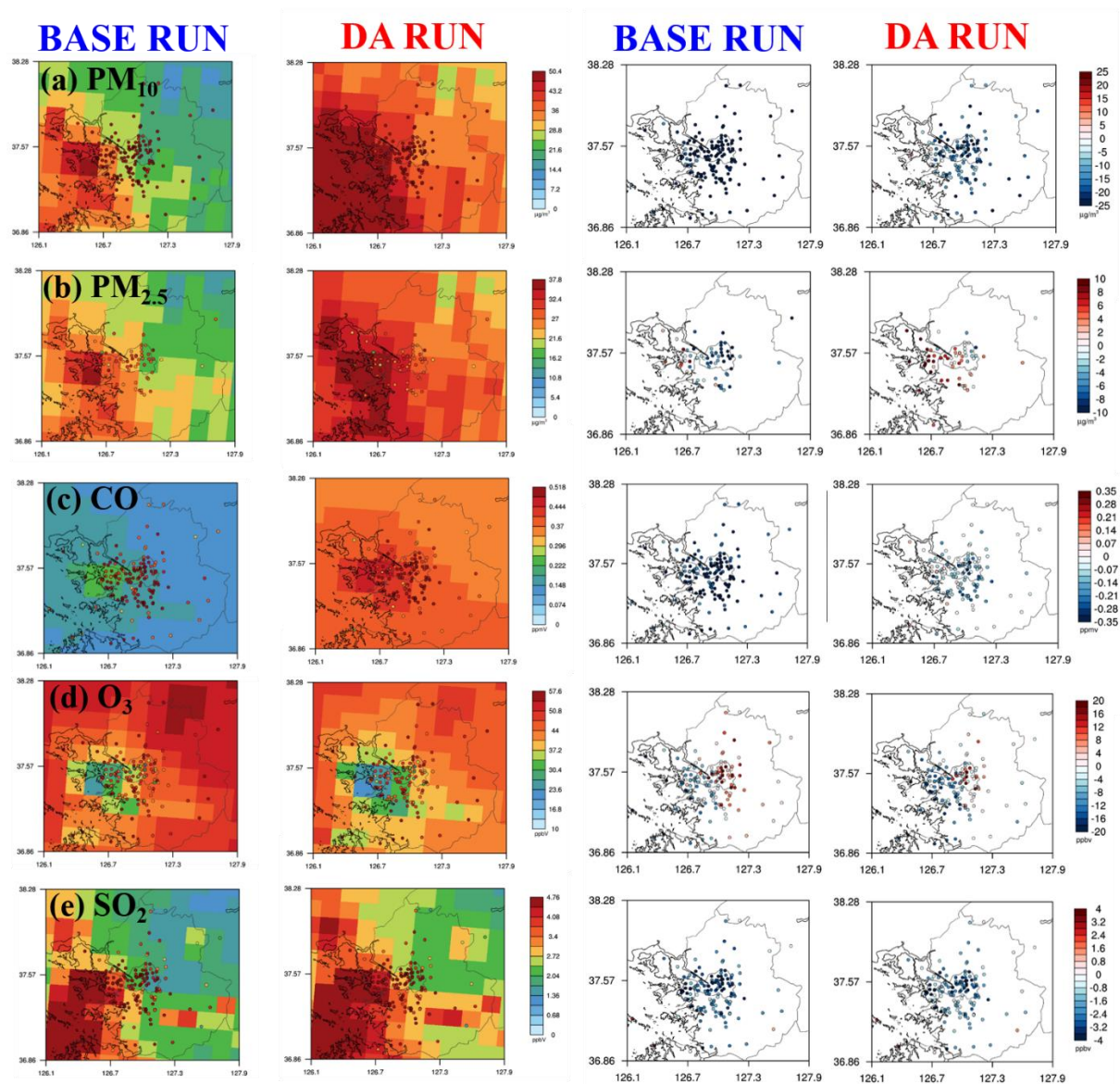


Figure 10. Spatial distributions (first and second columns) and bias (third and fourth columns) of (a) PM_{10} , (b) $PM_{2.5}$, (c) CO, (d) SO_2 , and (e) O_3 over Seoul Metropolitan Area (SMA) for the entire period of the KORUS-AQ campaign. Colored circles of first and second columns represent the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

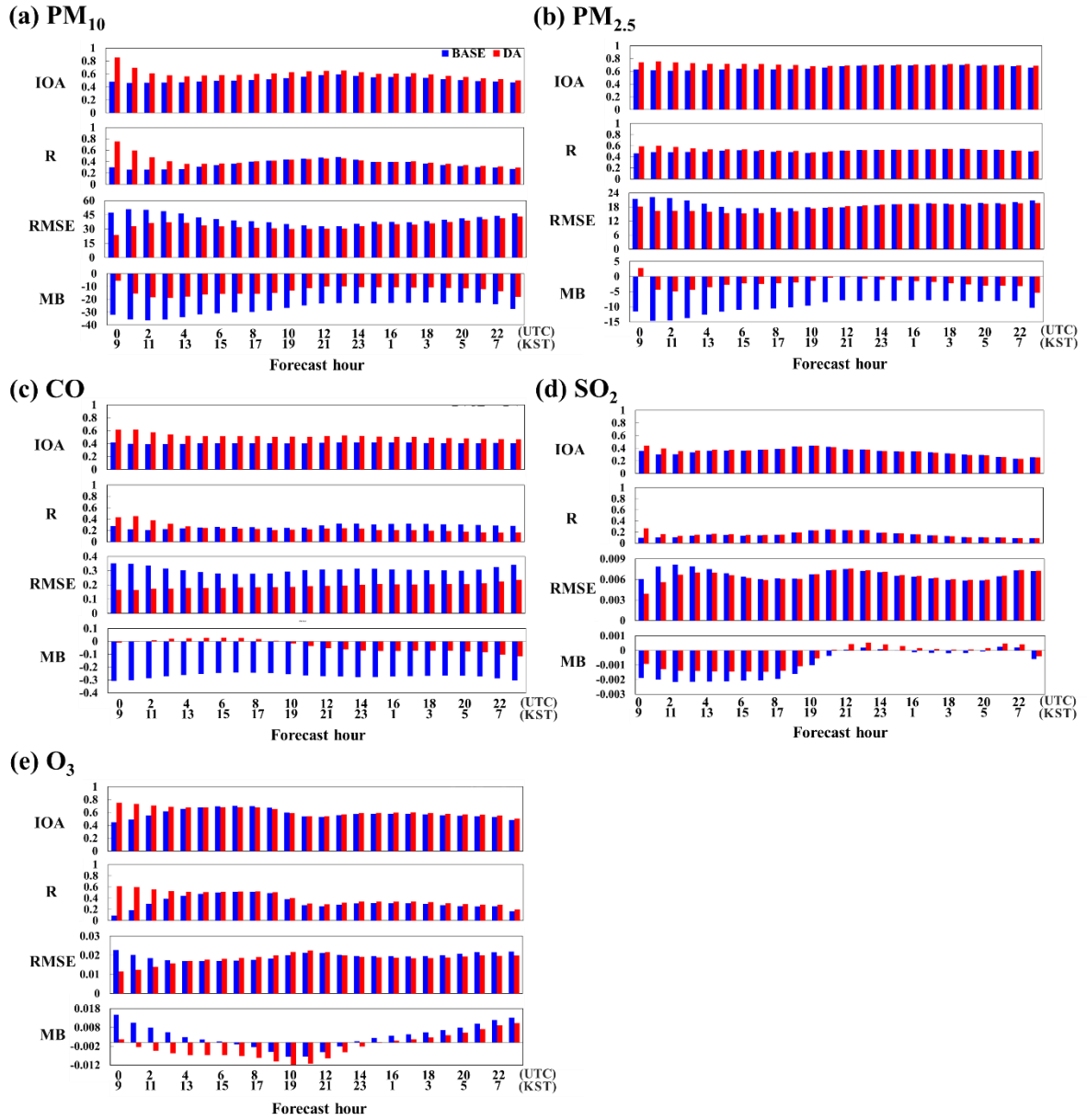


Figure 11. Time-series plots of four performance metrics (IOA, R, RMSE, and MB) for (a) PM₁₀, (b) PM_{2.5}, (c) CO, (d) SO₂, and (e) O₃ forecasts. The DA was conducted at 00:00 UTC. The units of RMSE and MB are $\mu\text{g}/\text{m}^3$ and ppmv for PM concentrations and for gaseous species, respectively. The definitions of the four performance metrics are shown in Appendix A.

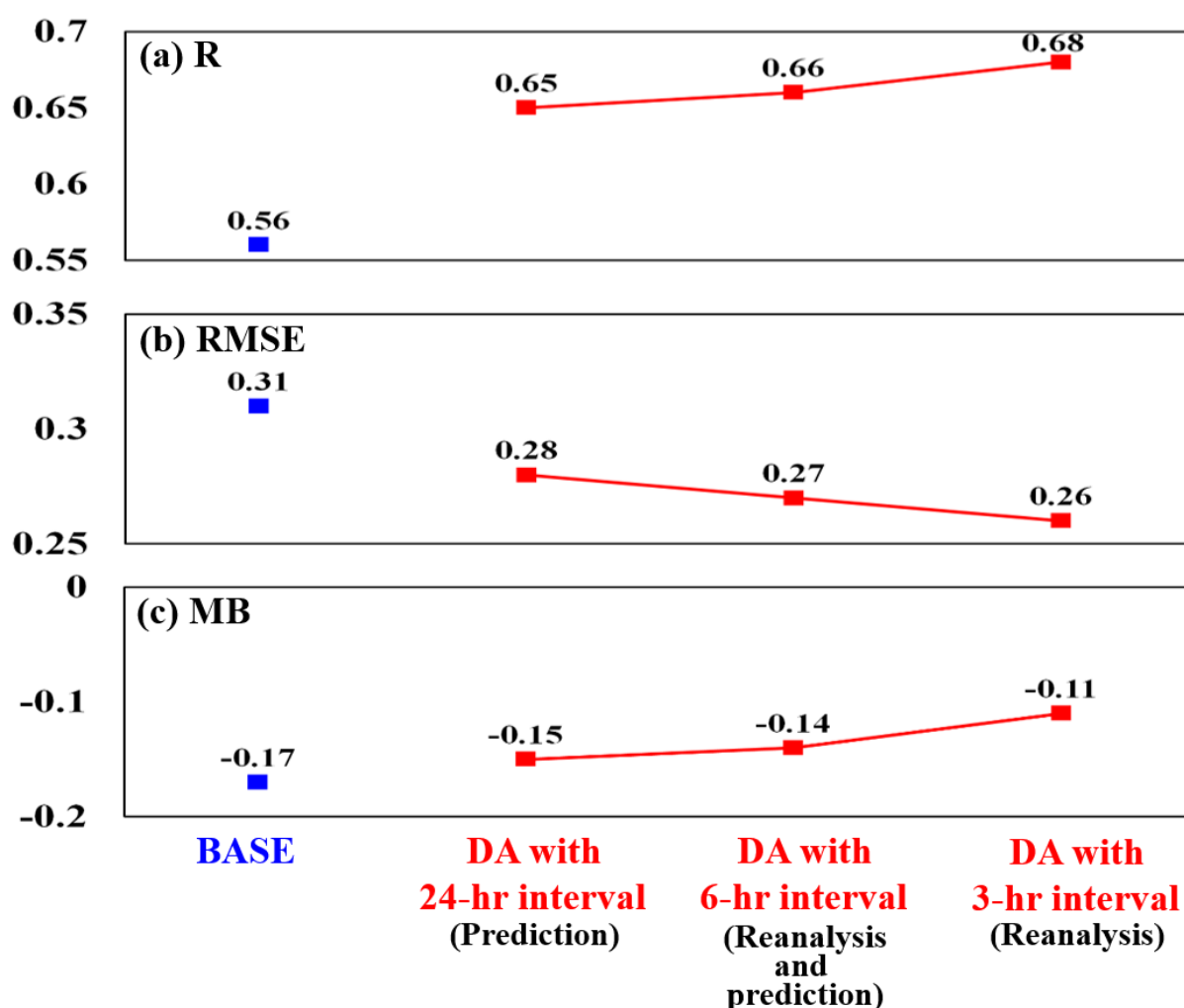


Figure 12. Variations of three performance metrics (R, RMSE, and MB) with time-intervals of data assimilations. For these tests, the GOCI AODs were used in the DA to update the initial conditions of the CMAQ model simulations. The results from the three CMAQ model simulations were compared with AERONET 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”.

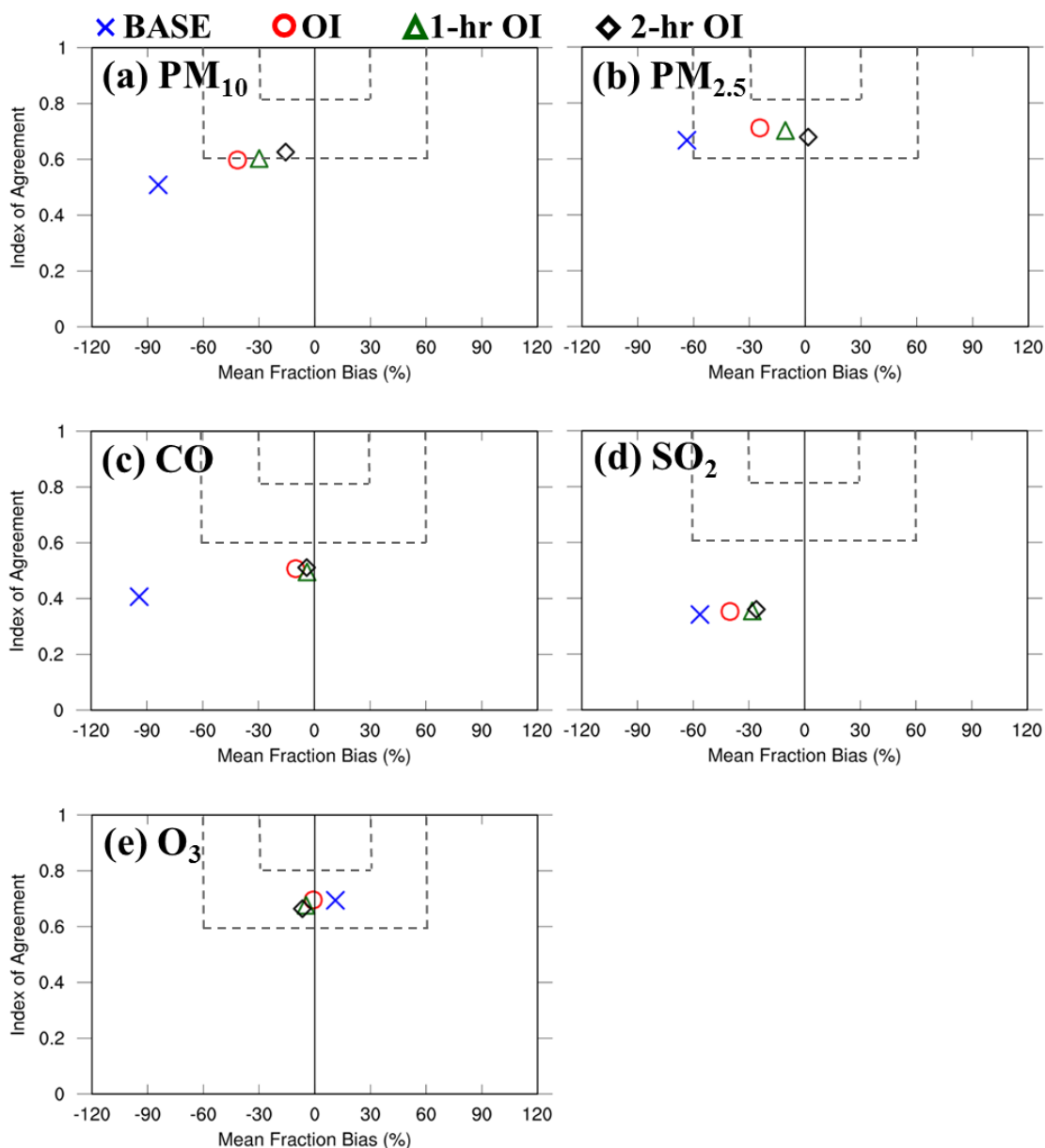


Figure 13. Soccer plot analyses for (a) PM₁₀, (b) PM_{2.5}, (c) CO, (d) SO₂, and (e) O₃. The CMAQ-predicted concentrations were compared with the Air Korea observations. Blue crosses, red circles, dark-green triangles, and black diamonds represent the performances calculated from the BASE RUN, the DA RUNs with the OI system, the 1-hour (hr) OI system, and the 2-hr OI system, respectively.

944 **Table 1.** Statistical metrics from BASE RUN and DA RUN with Air Korea observations over
945 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

946

947 **Table 2.** Statistical metrics from BASE RUN and DA RUN with Air Korea observations at
948 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

949