PLEASE NOTE

Reviewers' questions are in standard text.

Manuscript text is in *italic*.

Personal communication for reviewer is in **bold**.

Response to reviewer #1:

This study by Jeon et al. implemented a new hybrid Lagrangian-Eulerian model, STOPS, into CMAQ, to improve the air quality forecasting. Jeon et al. use the STOPS modeling framework with constrained PM from geostationary satellite AOD to improve the Asian dust event that occurred in South Korea on Feb 22-24, 2015. It demonstrates well how STOPS could be useful in air quality forecast, particularly for the unusual air quality events such as Asian dust transport. The merit of using STOPS is on low computational burden compared to CMAQ, which can be critical for emergency forecasting. The manuscript is well within the scope of GMD. However, the manuscript requires some revisions. Please see my comments below. In addition to those comment, I believe science writing in this manuscript should be improved, with focus on reducing the redundancy and increasing coherence within a paragraph. I have listed several places that need such improvement, but please try to improve throughout the manuscript (not limited to my list). When these comments/suggestions are addressed in the manuscript, I recommend this manuscript to be published in GMD.

The authors agreed with reviewer's comment about adding a detailed description of STOPS and in-line dust module in CMAQ v5.0.2, and additional meteorological evaluation results at each observational site. To that point, we have added a figure to briefly illustrate the basic concept of the STOPS model and also added an equation to better explain the in-line windblown dust module. Additionally, we have added the WRF evaluation results (statistics: RMSE, IOA and MBE) for all individual sites to depict a more comprehensive evaluation. Further, we shortened and revised the text so as to reduce redundancy, and have added comprehensive figures and tables for clarifying our results. Please see our responses to the specific comments.

Major Comments:

1. I encourage the authors to clarify the following point carefully throughout the manuscript. In my understanding, the STOPS model seems to be a great modeling tool, mainly due to less computational burden. It might be particularly useful when it needs to explore several possibilities. However, I don't think STOPS itself improves any air quality prediction. Also, the authors already stated that STOPS simulation results are relatively similar to CMAQ. I think the significant improvement in simulated PM10 was contributed by constraining PM10 based on GOCI AOD, not by using the STOPS model. CMAQ with the constrained PM10 from GOCI-AOD should also simulate a more accurate Asian dust. In short, I think STOPS does not contribute to "more accurate" forecasting but could help for "quicker" forecasting. If the authors agree with me, please change any relevant parts throughout the manuscript.

Thanks for the point. The authors agree that STOPS itself does not specifically improve any air quality prediction, but help for "quicker" forecasting. The significant improvement in the simulated PM$_{10}$ was contributed by constrained PM concentrations based on GOCI AOD. Thus, we revised all of the relevant parts and some sentences throughout the manuscript to avoid any possible misunderstanding from readers.

2. I suggest adding more detailed information of STOPS in Section 2.1. It is not easy to picture what exactly the STOPS model does (why is it a hybrid Lagrangian-Eulerian model?). I found the short description on the abstract (line 21-23) and the Figure 1 in Czader et al. (2015) quite helpful, which could be added to Section 2.1. Please clarify model domain and dispersion process used in STOPS: 1) does STOPS accounts for vertical and horizontal dispersion as it transport, like FLEXPART, which means it changes the number of grids carrying by STOPS over time?; 2) does STOPS carry a couple of grids in the defined STOPS domain or STOPS moves the defined STOPS domain over time (e.g., 61x61 gridcells in Section 4.1)?

As the reviewer suggested, we added a figure similar to Figure 1 in Czader et al., (2015) in the revised manuscript to show more detailed information of STOPS.

<Figure 1 in the revised manuscript>

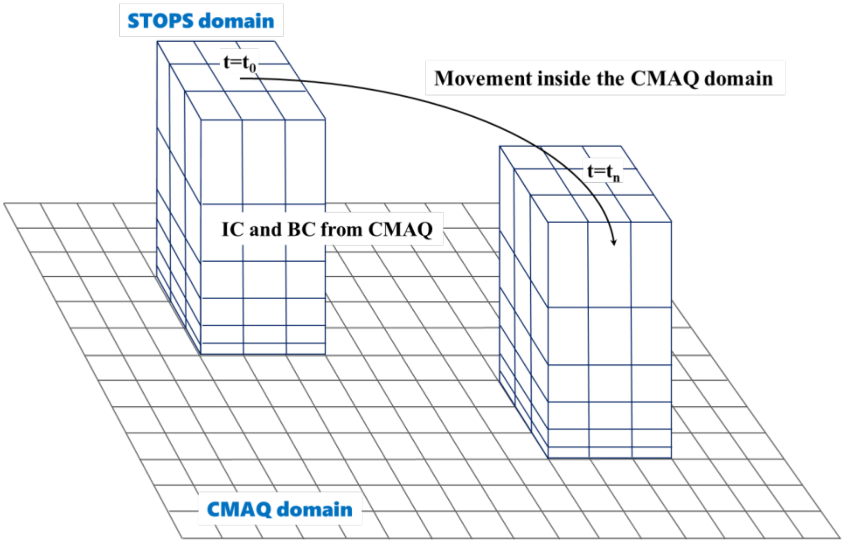


Figure 1. Conceptual diagram showing the basic structure and movement of the STOPS domain inside the CMAQ domain.

As shown in Figure 1 in the revised manuscript, STOPS has sub-domain inside the CMAQ domain and it moves along with the mean wind in its domain. The vertical structure and the physical and chemical process in STOPS are exactly same as in the host CMAQ model except for the calculation of advection fluxes. CMAQ uses horizontal wind velocity (u and v) from WRF to calculate horizontal advection fluxes; while STOPS calculates the difference between a cell horizontal wind velocity and the mean horizontal velocity in STOPS domain, so it can consider the moving speed and direction of STOPS domain for the calculation of advection fluxes. Since the STOPS domain moves over time, the horizontal velocity from WRF should be adjusted based on the movement of STOPS domain. Although STOPS is Eulerian-Lagrangian based model, it is close to Eulerian model rather than Lagrangian. STOPS is almost similar to CMAQ but has small domain size. The reason why STOPS is much faster than full CMAQ is that the number of grid cells in STOPS domain is much smaller than those in CMAQ domain. We revised section 2.1 in the manuscript by adding a figure and a couple of sentences for the better explain of STOPS model.

<Section 2.1 in the revised manuscript>

STOPS has the same vertical structure and simulates the same physical and chemical *processes as CMAQ, except for the calculation of advection fluxes. CMAQ uses horizontal wind velocity (u and v) from WRF to calculate horizontal advection fluxes, but STOPS calculates the difference between a cell horizontal wind velocity and the mean horizontal velocity in STOPS domain (Czader et al., 2015), so it can consider the moving speed and direction of STOPS domain for the calculation of advection fluxes. Since the STOPS domain moves over time, the horizontal velocity from WRF should be adjusted based on the movement of STOPS domain.*

3. I agree with the authors that the main reason for the PM10 underprediction in CMAQ is very likely missing dust emissions, as the threshold friction velocity calculation indicates. However, I don't agree with the authors on how to draw a conclusion that the model meteorology is accurate, mainly because the evaluation results, shown in Figure 3, are not comprehensive. Here are more specific questions related to the evaluation. First of all, why do the authors choose averaged values of 20 sites? I'd strongly prefer to see individual site evaluations. Alternatively, the individual site evaluation can be provided in supplementary material. Secondly, given that the long-range transport of Asian dust to influence South Korea, it is important to simulate correct meteorology from source regions to receptor regions. Would it be possible to include meteorological evaluations in Chinese source regions? Lastly, I encourage including more meteorological variables (such as precipitation, if there is any precipitation event during the event).

Firstly, we added WRF evaluation results (RMSE, IOA and MBE) for 20 sites (S1-S20) in the revised supplementary document as the reviewer suggested. Table S3 in the revised supplementary document shows evenly high IOA and low biases at 20 individual sites, indicating that the simulated meteorology over Korea (receptor regions) is reasonably accurate.

<Table S3 in the revised supplementary document>

Table S3. *Statistical parameters for the WRF simulation results during the entire simulation period (February 2015) at 20 observational sites. The location of each site is shown in Fig. 2 in the manuscript.*

Sites	*Temperature*	*Wind Speed*

	RMSE	*IOA*	*MBE*	*RMSE*	*IOA*	*MBE*
S1	*0.78*	*0.99*	*-0.08*	*1.12*	*0.97*	*0.03*
S2	*1.46*	*0.98*	*0.17*	*1.38*	*0.90*	*0.15*
S3	*2.49*	*0.90*	*-0.27*	*1.23*	*0.80*	*-0.85*
S4	*1.94*	*0.93*	*1.80*	*1.28*	*0.78*	*-0.21*
S5	*2.31*	*0.93*	*1.48*	*1.13*	*0.84*	*-0.40*
S6	*2.31*	*0.93*	*1.04*	*1.89*	*0.91*	*1.49*
S7	*2.48*	*0.96*	*-1.46*	*1.96*	*0.77*	*1.43*
S8	*2.58*	*0.93*	*-1.58*	*1.61*	*0.87*	*1.25*
S9	*1.40*	*0.94*	*1.39*	*1.19*	*0.86*	*1.12*
S10	*1.42*	*0.95*	*1.41*	*1.87*	*0.91*	*1.21*
S11	*2.02*	*0.97*	*-1.06*	*2.03*	*0.75*	*1.45*
S12	*2.70*	*0.78*	*-2.35*	*1.34*	*0.92*	*0.94*
S13	*2.11*	*0.94*	*1.24*	*1.24*	*0.88*	*0.85*
S14	*1.59*	*0.95*	*1.01*	*2.07*	*0.93*	*1.46*
S15	*2.67*	*0.89*	*-2.29*	*2.37*	*0.76*	*1.90*
S16	*1.39*	*0.98*	*0.43*	*1.59*	*0.89*	*0.90*
S17	*2.48*	*0.84*	*-1.71*	*1.98*	*0.74*	*1.36*
S18	*1.60*	*0.96*	*-1.09*	*2.64*	*0.72*	*1.27*
S19	*1.58*	*0.95*	*1.17*	*2.03*	*0.82*	*1.02*
S20	*1.12*	*0.96*	*0.98*	*1.59*	*0.89*	*0.90*
Average	*1.92*	*0.93*	*0.01*	*1.68*	*0.85*	*0.86*

Secondly, the authors agree that the meteorology in source regions (China and Mongolia) is also important for the simulation of long-range transport of Asian dust; hence the data need to be evaluated. This study showed accurate meteorology only in receptor regions (Korea) not in source regions (China and Mongolia) due to the limited availability of the data. As the reviewer indicated, uncertainty in meteorology (particularly in source regions) could be one of possible reason for the PM_{10} underestimation. We have added the requisite description regarding the uncertainty in meteorology in section 3.1 in the revised manuscript.

<Section 3.1 in the revised manuscript>

As shown in Fig. 4 and Table S3, meteorological fields such as temperature and wind speed over receptor regions (Korea) showed close agreement with observations, even during the Asian dust period. It suggests that the underestimated PM_{10} *concentration was likely due to the uncertainty in meteorology over source regions (China and Mongolia), and/or faulty* *estimation of dust emissions for the CMAQ simulation. We attributed the main reason for the* PM_{10} *underestimation to poorly estimated dust emission because CMAQ showed poor performance only during the Asian dust event days.*

Finally, there were several challenges in obtaining observational data in China and Mongolia. Also, the surface data for this study provided only temperature and wind variability. For these reasons we could not include meteorological evaluations in source regions and evaluation results for other factors such as precipitation.

4. Please provide a brief description of the CMAQ dust emission parameterizations used in your forecast modeling. It will help readers to understand what the underpredicted threshold friction velocity affects to dust emissions.

As the reviewer suggested, we provided a brief description of the CMAQ in-line windblown dust module as shown below.

<Section 3.1 in the revised manuscript>

The module calculates the vertical dust emission flux (F) by following formula described by Fu et al. (2014).

$$F = \sum_{i=1}^{M}\sum_{j=1}^{N} K \times A \times \frac{\rho}{g} \times S_i \times SEP \times u_* \times ({u_*}^2 - {u_{*ti,j}}^2)$$

where i and j represent the type of erodible land and soil, K is the ratio between vertical and horizontal flux, A is the particle supply limitation, ρ *is the air density, g is the gravitational constant,* S_i *is the area of the dust source,* SEP *is the soil erodible potential,* u_* *is the friction velocity, and* $u_{*ti,j}$ *denotes the threshold friction velocity.*

Also, we added a sentence describing the importance of threshold friction velocity on the calculation of dust emission flux to better explain the reason for the underestimated dust emission from the CMAQ in-line module.

<Section 3.1 in the revised manuscript>

Several studies (e.g. Choi et al., 2008; Fu et al., 2014) have reported that the threshold friction velocity plays a key role in the calculation of dust emission flux because the threshold *can determine the possibility of the lifting of dust particles.*

Minor comments:

1. <Title>: I'd suggest changing a title. What about "Computationally efficient air quality forecasting tool: implementation of a hybrid Lagrangian-Eulerian model into CMAQ v5.0.2"?

The authors agreed to change the title as the reviewer suggested. However, we added a phrase "for a prediction of Asian dust" to emphasize that this is a case study for an Asian dust event. Also, we used "STOPS model" instead of "a hybrid Lagrangian-Eulerian model", because we thought "a hybrid Lagrangian-Eulerian model" is too generic to be used in the title. In conclusion, we changed the title of this study as "Computationally efficient air quality forecasting tool: implementation of STOPS model into CMAQ v5.0.2 for a prediction of Asian dust".

2. <Abstract> : I'd strongly suggest re-writing this section. Overall abstract seems to sound quite redundant. Please consider taking the suggestions below.

Page 1; line 17-19 – Please consider moving this to the end of Abstract and either delete or modify this phrase ("for a more accurate prediction of Asian dust event in Korea"): see the major comment above.

Page 1; line 20-21 – I'd suggest deleting sentence. It is mentioned in line 31-33.

Page 1; line 24-27 – Please consider deleting this as well. Next a few sentences basically say the same information. Having this sentence, it sounds too redundant.

Page 1; line 29-31 – I'd suggest modifying this. The following is my suggestion: "The underestimated PM10 concentration is very likely due to missing dust emissions in CMAQ rather than incorrectly simulated meteorology as the model meteorology agrees well with the observations."

Page 1; line 32 – Please delete "we use the STOPS modeling system inside the CMAQ model, and", and please modify "we run several STOPS simulations using" to "we used the STOPS model with".

Page 2; line 2-4 – Please shorten the sentence. "The simulated PM10 from the STOPS simulations were improved significantly and closely matched to surface observations".

Page 2; line 5-9 – Please see my major comment 1.

We re-wrote the Abstract section based on the reviewer's comments. We shortened and changed the sentences as the reviewer suggested and deleted unnecessary sentences to reduce the redundancy.

<Abstract in the revised manuscript>

Abstract. *This study suggests a new modeling framework using a hybrid Lagrangian-Eulerian based modeling tool (the Screening Trajectory Ozone Prediction System, STOPS) for a prediction of an Asian dust event in Korea. The new version of STOPS (v1.5) has been implemented into the Community Multi-scale Air Quality (CMAQ) model version 5.0.2. The STOPS modeling system is a moving nest (Lagrangian approach) between the source and the receptor inside the host Eulerian CMAQ model. The proposed model generates simulation results that are relatively consistent with those of CMAQ but within a comparatively shorter computational time period. We find that standard CMAQ generally underestimates* PM_{10} *concentrations during the simulation period (February 2015) and fails to capture* PM_{10} *peaks during Asian dust events (22-24 February, 2015), The underestimated* PM_{10} *concentration is very likely due to missing dust emissions in CMAQ rather than incorrectly simulated meteorology as the model meteorology agrees well with the observations. To improve the underestimated* PM_{10} *results from CMAQ, we used the STOPS model with constrained PM concentrations based on aerosol optical depth (AOD) data from Geostationary Ocean Color Imager (GOCI), reflecting real-time initial and boundary conditions of dust particles near the Korean Peninsula. The simulated* PM_{10} *from the STOPS simulations were improved significantly and closely matched to surface observations. With additional verification of the capabilities of the methodology on concentration estimations and more STOPS simulations for various time periods, STOPS model could prove to be a useful tool not just for the predictions of Asian dust but also for other unexpected events such as wildfires and upset emissions events.*

3. <1. Introduction>

Page 2; line 18-21 - I'd suggest changing "Severe PM events … Gobi Desert" to "Dust emissions from Mongolia and Gobi Desert".

Page 2; line 23 – please change “become” to “are”.

Page 2; line 29 – Please rephrase “the numerous factors such as meteorology and emissions ... PM concentrations”. It sounds a bit unclear.

Page 2; line 21 – Add “modeling” in front of “studies”; change “described” to “shown” and delete “simulation”.

Page 3; line 31 to Page 3; line 9 – This paragraph should be rewritten in order to deliver the key point clearly, which, I think, improving meteorology and emission inventory do not help better Asian dust forecasting due to the uncertainty in dust emission modeling. Besides, please delete the last sentence (Therefore, ~): the first part is too obvious to mention, and the second part is somewhat debatable (especially “primarily”) and contradicts with “accurate meteorology” above.

Page 3; line 25 – This “(STOPS, hereafter)” should be moved above, where STOPS is mentioned in the first time.

Page 3; line 22-35 – I found this paragraph This paragraph doesn’t sound coherent. Please use present tense to state goals and objectives and past tense for methods. Please also modify the paragraph based on my major comment 1. It is incorrect to say that STOPS enhance the PM predictions.

Page 3; line 23 – Delete “simulated”; add “to” in front of “determine”.

Page 3; line 24 – Delete “particularly”, as this study focuses on Asian dust event only.

We revised the Introduction section based on the reviewer’s comments. We re-wrote some sentences more clearly and removed a couple of unnecessary sentences as the reviewer suggested.

4. <2.2 Modeling system and experimental design>

Page 5; line 4-5 – I think this sentence fits better in the end of next paragraph.

Page 5; line 10 – why do you mean by “refer to the CAPPS emissions”?

Page 5; line 18 – delete “for the simulation”

Page 5; line 18-23 – Please shorten the sentences.

Page 5; line 24 – Please remove “listed in Table 1” and list the date here.

We shortened, moved and deleted some sentences in section 2.2 (in the revised manuscript) as the reviewer suggested.

5. <2.3 In-situ and satellite measurements>

Page 5; line 29 – “referred to” to “use”

Page 5; line 36 – what is this “500 m resolution” for? Why is it different from AOD’s 6 km resolution?

Page 6; line 1 – “550 nm AOD” to “AOD at 550nm”

500 m and 6 km are the resolutions of original GOCI data and retrieved one by Choi et al. (2016) algorithm. The retrieved GOCI data with a 6 km resolution were used in this study. We corrected two phrases in section 2.3 (in the revised manuscript) as the reviewer suggested.

6. <3.1 Comparison with surface measurement>

Page 6; line 20-22 – Please define RMSE, IOA and MBE and explain what each measure indicates briefly.

Page 6; line 26-29 – Please see the major comment 3.

Page 6; line 30-36 – CMAQ dust emission modeling should be explained before this result. Please add the brief description in method section.

As the reviewer suggested, we added brief description of the statistical parameters used in this study (RMSE, IOA and MBE) in section 2.3 (in the revised manuscript).

<Section 2.3 in the revised manuscript>

The following statistical parameters were used for the evaluation of the performance of WRF and CMAQ simulations: Index Of Agreement (IOA), Mean Bias Error (MBE) and Root Mean Square Error (RMSE). These are defined as:

$$IOA = 1 - \frac{\sum_{i=1}^{N}(P_i - O_i)^2}{\sum_{i=1}^{N}(|P_i - \bar{P}| + |O_i - \bar{O}|)^2}$$

$$MBE = \frac{\sum_{i=1}^{N}(P_i - O_i)}{N}$$

$$RMSE = \sqrt{\frac{\sum_{i=1}^{N}(P_i - O_i)^2}{N}}$$

where N is number of data points and P_i *and* O_i *denote CMAQ-simulated and observed concentrations, respectively.*

We also added the WRF evaluation results at individual sites in Table S3 (in the revised supplementary document), and a formula for dust estimation used in CMAQ in-line dust module in section 3.1 (in the revised manuscript) for the better explanation. Please see our responses for question 3 and 4.

7. <3.2 Comparison with satellite-based observation>

Page 7; equations 4-6 – It looks like empirically derived method. Does the method by Roy et al. (2007) tested over the Korea as compared to more theoretical-based (Mie theory) optical properties? Is it reasonable to use it for Korea? Also, why isn't there no water uptake by organic aerosol [OM] in Eq 5?

Figure 4 – It is good that the CMAQ AOD field shows removed areas with GOCI bad pixels. However, it would be also helpful to present CMAQ AOD without removing any areas in the supplementary materials. It could show what GOCI might miss in those areas.

Page 7; line 32 – delete "the same results"

Page 7; line 34 – Do you actually mean "PM precursor" or "PM and its precursor"? If it is indeed specifically "PM precursor", please provide further explanation. Next sentence about meteorology should be re-considered (see major comment)

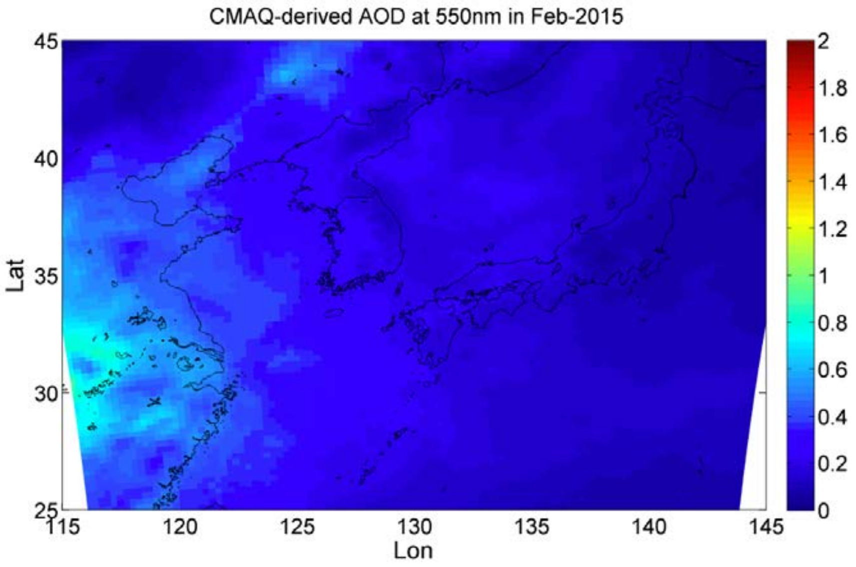
Page 8; line 3 - please add year: Feb 22-24, 2015. Please make the same corrections throughout the manuscript, if possible.

Page 8; line 15-16 – please change "the high amounts of dust particles" to "the high dust concentrations".

Page 8; line 19-20 – This should be modified with my major comment 1. I'd suggest changing to this: "We use STOPS to explore how to improve PM10 simulation."

Firstly, the empirical method used in this study has successfully been tested in East Asia (Park et al., 2011; Song et al., 2008) which is preferred to the Mie theory in this region. This is mainly because of the fact that aerosols properties including size distribution have not been precisely characterized in this region to allow us to use the Mie-theory extinction coefficient calculations. This issue was partly discussed in two mentioned papers. The OM hygroscpocitiy are highly uncertain, and to best of our knowledge it has not been parameterized yet. It should be mentioned that the significant portion of dust particles is NH_4NO_3 and SO_4^{2-}, therefore OM concentrations are not strongly prominent.

Secondly, as the reviewer suggested, we made a figure showing the CMAQ AOD without masking of GOCI bad pixels (Please see the figure shown below). However, it does not entirely match with previous figure (Figure 5-(b) in the revised manuscript) because bad pixels in GOCI were not filtered out for the calculation of monthly mean AOD from CMAQ. Although the below figure shows the CMAQ-derived AOD over whole areas in the modeling domain, it cannot be directly compared with figures in Figure 5 (in the revised manuscript). For this reason, the authors decided not to add the below figure to the supplementary document to avoid unnecessary argument.



Lastly, we revised all the sentences in section 3.2 as the reviewer suggested.

8. <4.2 PM_{10} forecasting using STOPS>

Page 9; line 6-8 – This sentence is unnecessarily long. Please remove “that is, the … failed”.

Page 9; line 8-9 – This should be rephrased, esp. “the most recent and accurate input data”. It makes me think about meteorology, emissions, initial and boundary conditions. If the constrained PM10 derived from GOCI AOD is only read in the first time, it is considered initial concentration and thus “input data”. However, the way you used the constrained PM10 derived from GOCI AOD in Section 4.2.2 seems more than initialization and close to nudging.

Page 9; line 13-18 – Please remove this part. This is out of placed and doesn’t have much new information, in my opinion. If the authors want to make a point that the CMAQ with constrained PM using GOCI AOD is less desirable as a forecasting tool due to their long simulations, perhaps do it elsewhere (maybe the end of the paragraph).

Page 9; line 18 – what do you mean by “dust core”? center of dust storm?

Page 9; line 26- do you actually mean “on the STOPS domain”? Perhaps it is “on the STOPS results”? Also, perhaps “would be diminished” is better than “would be mitigated”?

We removed and revised a couple of unnecessary sentences and confusing phrases as the reviewer suggested (Section 4 in the revised manuscript).

9. <4.2.1 Satellite-adjusted PM concentrations>: This section is particularly confusing. Please re-write them and use figure or diagram to help readers to understand the method.

Page 9; line 31 – Please remove “To provide ~ AOD into account,” and clarify “at the beginning of the updated forecast”.

Page 9; line 34 – Perhaps “as a constraint” is correct?

Page 10 – Isn’t the second paragraph better to move?

Page 11; line 2-3 – Fix line break 4.2.2.

As the reviewer suggested, we re-wrote section 4.1 (in the revised manuscript) to better explain the method we used for PM constraining. We added a figure, which briefly describes entire procedures of the new PM forecasting using STOPS with GOCI-derived AOD data.

<Figure S3 in the revised supplementary document>

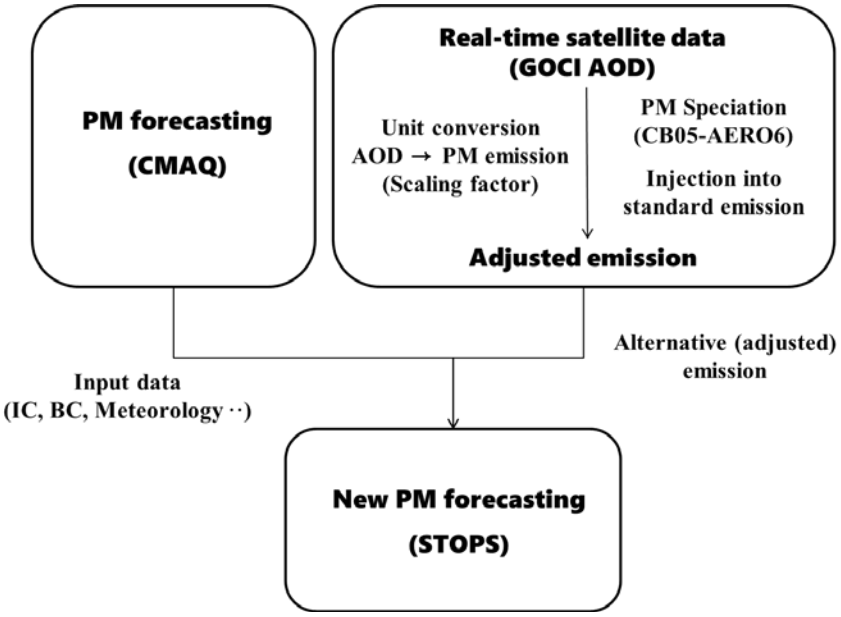


***Figure S3**. Schematic flowchart describing the procedures of the new PM forecasting by STOPS using the real-time AOD data from GOCI.*

10. <4.2.2 Enhanced PM10 forecasting using STOPS>

Page 11; line 22 - why did you said “were assumed to”?

Page 11; line 29-30 – please shorten to “Figure 8 exhibits clear…”

Page 11; line 27 – please add “, shown in Fig. 8,” after using STOPS

Page 11; line 32 – please change to “because of the poor dust emission modeling in CMAQ”.

Page 11; line 36~ - Isn’t this already mentioned in Line 30?

Page 12; line 32 – Remove “changed” in “To verify the changed horizontal”

We removed some unnecessary sentences, and revised all of the addressed phrases and sentences as the reviewer suggested in order to reduce redundancy (Section 4.2 in the revised manuscript).

11. <Summary>: Please revise the summary section if it is subject to the major comments.

Page 13; line 22 – “but with” to “but used”

Page 13; line 24 – add comma between “dust events” and “we”

We revised the Summary by considering all of the changes in each section in the revised manuscript.

12. <Table & Figures>

Table 2 – “Without Dust Events” to “Without dust events”

Figure 1 – It is hard to find the site location. I was able to find only 17 sites. Can you use color symbol for sites?

Figure 2 – It would be nice, if the dust event days were shown in the figure.

Figure 6 – Does white space shown in the map represent for very low AOD or does it also include areas with missing pixels? Just in cases missing areas should be shown in white.

Figure 7 – Please double check the caption. It says standard and constrained CMAQ runs, while “constrained CMAQ run” is never discussed in the main text.

We corrected “Without Dust Events” in Table 2 (in the revised manuscript) to “Without dust events”, changed Figure 2 (in the revised manuscript) by adding 3 missing sites and using color symbols, and marked the Asian dust event days in Figure 3 (in the revised manuscript). Also, we revised caption in Figure 7 and 8 (in the revised manuscript) for the better explanation.

<Figure 2, 3, 7 and 8 in the revised manuscript>

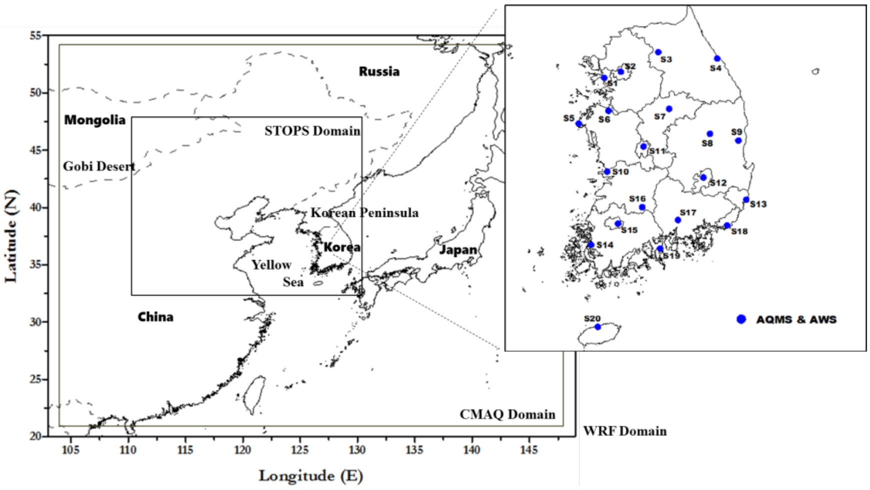


Figure 2. *Domains for the WRF, CMAQ and STOPS modeling. The right panel shows the location of the air quality monitoring stations (AQMS) and automatic weather system (AWS) sites used in this study.*

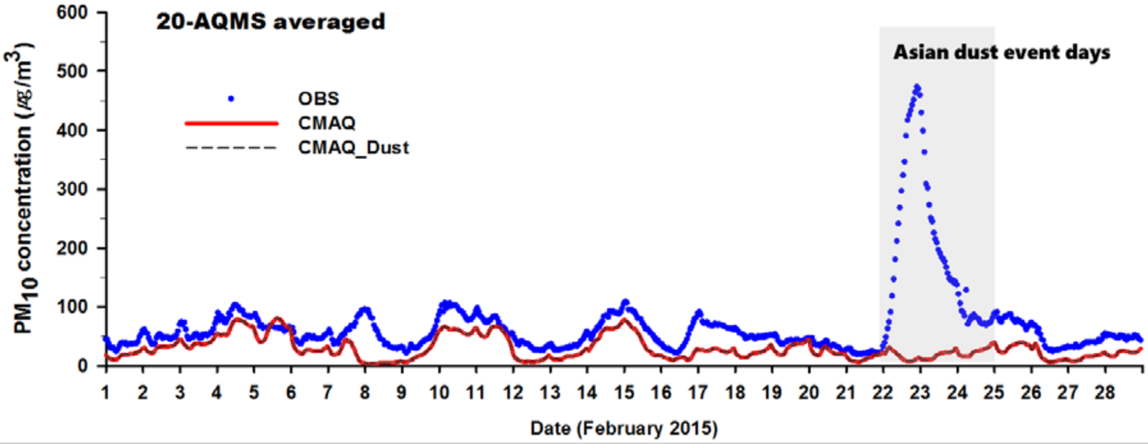


Figure 3. *Time series of observed (OBS, blue dots) and simulated (CMAQ: red line, CMAQ_Dust: black dashed line) PM_{10} concentrations in February 2015. The values are averaged values for 20 AQMS sites: CMAQ_Dust is closely coupled with the standard CMAQ modeling results (red line).*

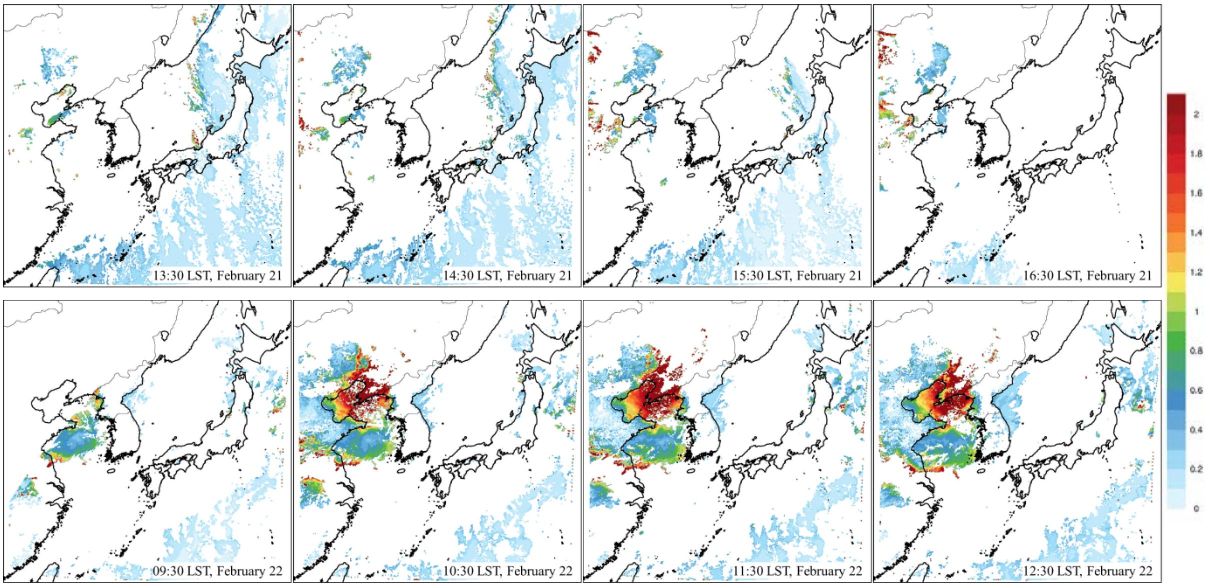


Figure 7. *The GOCI-derived AOD (550 nm) from 13:30 LST on 21 February to 12:30 LST on 22 February in 2015. The white-colored areas represent missing pixels.*

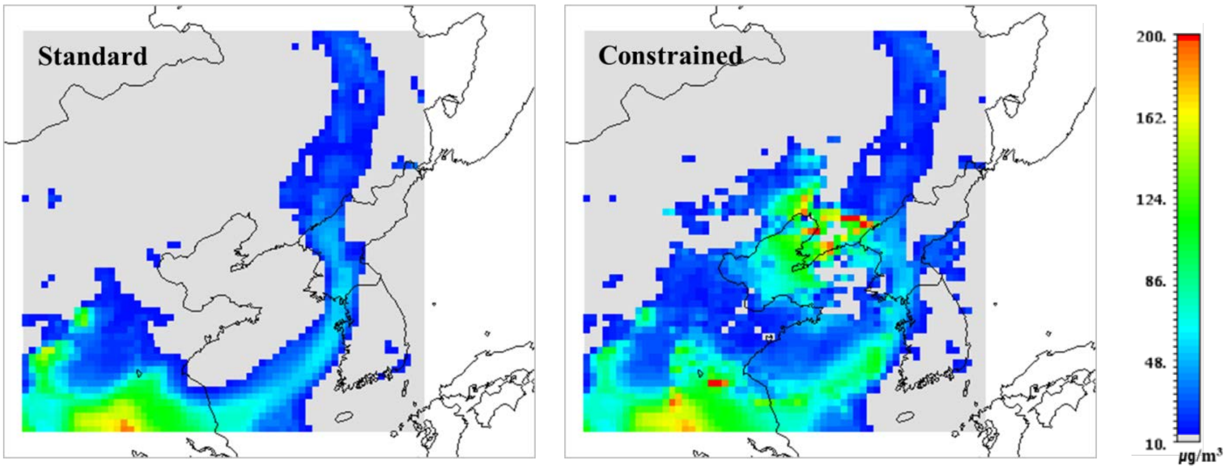


Figure 8. *Difference of the simulated* PM_{10} *concentrations* ($\mu g\ m^{-3}$) *between the standard CMAQ run (left) and STOPS forecasting run with alternative emission estimated according to GOCI-derived AOD (right) inside the STOPS domain at 12:00 LST on 22 February in 2015.*

PLEASE NOTE

Reviewers' questions are in standard text.

Manuscript text is in *italic*.

Personal communication for reviewer is in **bold**.

Response to reviewer #2:

The authors agreed with reviewer's comment regarding the necessity of the model configurations for WRF and CMAQ simulations, synoptic weather chart in the Asian dust event day, detailed description of in-line dust module in CMAQ v5.0.2, and more clear explanation of the methodology we used for STOPS forecasting. We added a couple of figures and tables, and additional description for them for better understanding, and revised a lot of sentences based on the reviewer's suggestion to reduce redundancy. Also, we revised a couple of confusing and misleading paragraphs in the manuscript with the professional English editing and proof reading to make the manuscript more concise and readable.

Again, the authors responded to most of the reviewer's comments and strengthened our revised manuscript and supplementary document. Please see our responses to the specific comments.

Specific Comments:

1. P3, line 24-35, grammatical errors. For describing what was done in this paper, the past tense would be used. Not just in this paragraph, many grammatical errors are in the text. Sentences are not conveying arguments smoothly that I need to read them a few times to understand authors' intention (such as P3, line 30-33). Sometimes, the wordings are redundant in carrying out the arguments (like p7, p9 line 5-10, p9, line 13-24). With the help of professional English editing and proof reading, the manuscript will be more concise and readable.

The authors revised all of the confusing and misleading paragraphs throughout the manuscript with the professional English editing and proof reading to make the

manuscript more concise and readable.

2. P3, line 11, give citation (Byun and Schere, 2006) when the model is 1st mentioned in the paper.

We added a citation, "Byun and Schere, 2006", in the sentence.

3. P3, line 27, "We utilized STOPS: : :",
P3, line 29, "input data inside the modeling domain."

We corrected the sentences as suggested by the reviewer.

4. P4, line 5, re-phase the sentence to C1 "A small sub-domain of STOPS was configured inside the CMAQ domain and it moves along with the mean wind from CMAQ."

We revised the sentence as the reviewer suggested, and added a figure in the revised manuscript for the better understanding from readers.

<Figure 1 in the revised manuscript>

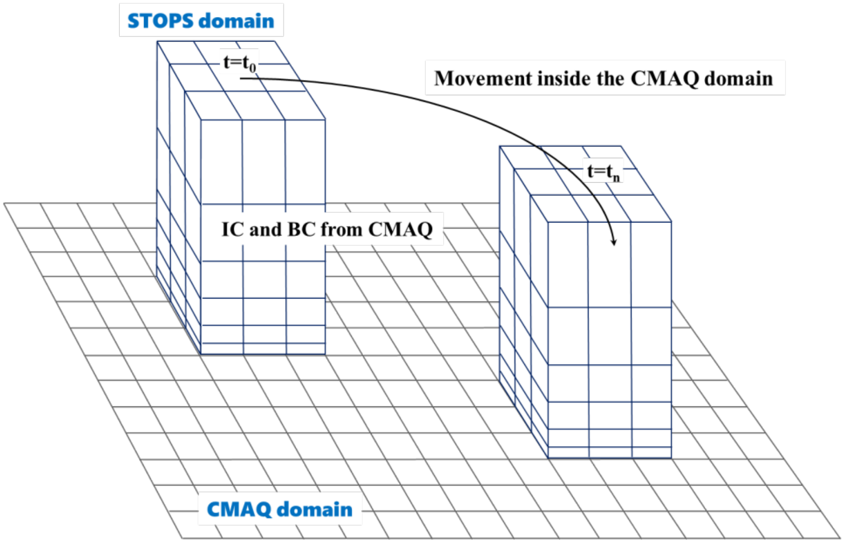


Figure 1. Conceptual diagram showing the basic structure and movement of the STOPS domain inside the CMAQ domain.

5. P4, line 9, the sentence is confusing, please rewrite it.

We re-wrote the sentence to clearly explain how STOPS calculates advection fluxes.

<Section 2.1 in the revised manuscript>

STOPS has the same vertical structure and simulates the same physical and chemical processes as CMAQ, except for the calculation of advection fluxes. CMAQ uses horizontal wind velocity (u and v) from WRF to calculate horizontal advection fluxes, but STOPS calculates the difference between a cell horizontal wind velocity and the mean horizontal velocity in STOPS domain (Czader et al., 2015), so it can consider the moving speed and direction of STOPS domain for the calculation of advection fluxes. Since the STOPS domain moves over time, the horizontal velocity from WRF should be adjusted based on the movement of STOPS domain.

6. P4, line 10-11, ": : :is determined by the layer-averaged wind from the 1st model up to the top of planetary boundary layer (PBL), weighted by the layer thickness.",

P4, line 27, "but in this study, STOPS has been updated to v1.5 and implemented in CMAQ v5.0.2.",

P4, line 31-33, No need to give citation again for the CMAQ. "In this study, we configured the CMAQ model with a domain in a grid resolution of 27 km covering the northeastern part of Asia: : :"

We revised the sentences as the reviewer suggested.

7. P4, line 29, the list and description of all the simulations – standard CMAQ, CMAQ with windblow dust, CMAQ with adjusted emission and four STOPS with adjusted emission are expected in the section titled as experimental design. It can be in its own section if appropriate.

We changed the title of section 2.2 from "*2.2. Modeling system and experimental*

***design*” to “*2.2. Modeling system*” because the section does not include any experimental procedure. We have included the descriptions of each simulation (CMAQ and STOPS) in their relevant sections to better explain the methodology, data and options used for each simulation case.**

8. P5, line 1-2, “Gobi Desert which is a major source of Asian dust.”

We corrected the sentence as suggested by the reviewer.

9. P5, line 2, spell out full name of “CB05” and “AERO6” and provide citations.

We added the full names and citations for them in the revised manuscript.

<Section 2.2 in the revised manuscript>

The Carbon Bond chemical mechanism (CB05) (Yarwood et al., 2005) and the AERO6 aerosol module (Nolte et al., 2015) were used for gas-phase and aerosol chemical mechanisms, and initial and boundary conditions were obtained from the standard CMAQ profile.

10. P5, line 5-22, missing CMAQ and WRF’s model configuration. Please list physics options used in WRF and the schemes (such as advection, deposition, etc: : :) used in CMAQ. Also, the model configuration for STOPS should be described in this section.

We added model configurations for WRF and CMAQ simulations in the revised supplementary document. Also, we moved section 4.1 (Configuration of STOPS) to this section as the reviewer suggested.

<Table S1 and S2 in the revised supplementary document>

***Table S1**. Configuration and detailed physical options for WRF simulation*

Number of grids	*181 × 143*

Horizontal resolution	*27 km*
Vertical layers	*33 layers*
Initial data	*1°× 1° NCEP Final Operational Global Analysis (FNL)*
Microphysics option	*WSM 3-class simple ice scheme*
Radiation option	*RRTM (long wave) / Dudhia (short wave) scheme*
Surface layer option	*Monin-Obukhov (Janic Eta) scheme*
Land-surface option	*Unified Noah land-surface model*
PBL option	*YSU scheme*
Cumulus option	*Kain-Fritsch (new Eta) scheme*

Table S2. *Same as Table S1, but for CMAQ*

Meteorology	*WRF*
Number of grids	*174 × 128*
Horizontal resolution	*27 km*
Vertical layers	*15 layers*
Chemical mechanism	*CB05 (gas-phase) / AERO6 (aerosol)*
Chemical solver	*Smvgear*
Horizontal advection	*Yamo*
Horizontal diffusion	*Multiscale*
Vertical advection	*WRF*
Vertical diffusion	*ACM2*
Deposition	*M3dry*
Anthropogenic emissions	*MIX-2010 / CAPSS 2011*
Dust emission model	*In-line windblown dust model*

11. P5, line 24, please provide overview of the synoptic weather pattern during the dust event that will help readers to interpret the model result.

We added two synoptic weather charts in the revised supplementary document to show the synoptic weather pattern on the first day of the Asian dust event (22 February, 2015), which resulted in the transport of massive dust from Mongolia region to the Korean Peninsula.

<Figure S1 in the revised supplementary document>

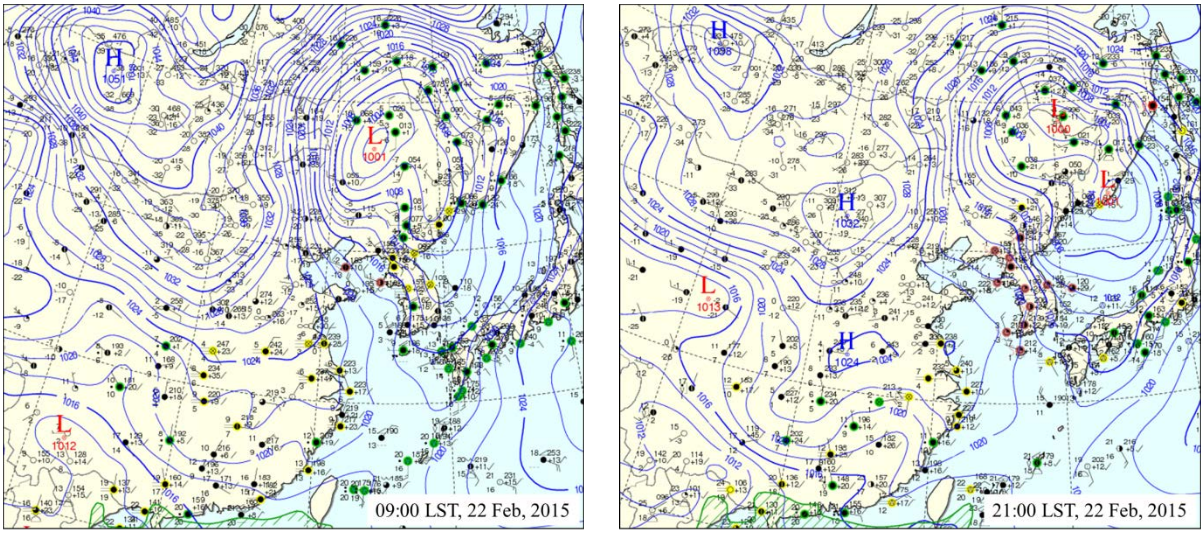


***Figure S1**. Surface-level synoptic weather chart near the Korean Peninsula on 22 February in 2015, which is the first day of the Asian dust event in this study.*

<Section 2.2 in the revised manuscript>

During the event days, massive dust over the GOBI desert and Mongolia region was transported to the Korean Peninsula. This happened due to the southeastward wind resulting from high pressure over the Mongolia region and low pressure over the northeastern part of China (Fig. S1 in the supplementary document).

12. P5, line 23-25, the paragraph should be re-written to give clear information about the simulation period and when the dust event happened. “The WRF-CMAQ simulations were conducted for the period of January 21st – February 28th, 2015 which included the first ten days for spin-up. Evaluations applied to the month of February, 2015 and the three-day Asian dust event occurred during February 22nd – 24th. The PM surface observations measured at

the surface stations in Korea are listed in Table 1.

We re-wrote the paragraph as the reviewer's suggested.

13. P5, line 29, "This study used surface observational data: : :"

We revised the sentence as suggested by the reviewer.

14. P6, line 3, what does it mean for constraining of PM concentration? Is it through data assimilation? If so, it should be described in methodology section like 2.2.

We did not use data assimilation technique for constraining PM concentration in forecasting. Usually, the data assimilation techniques are computationally more expensive than the simplified constraining approach. As described in section 4.1 in the revised manuscript, we regarded the GOCI-derived AOD as a surrogate for PM emissions and hence indirectly constrained the original PM concentrations by using the alternative emissions. The GOCI-derived AOD was converted to emission unit and the converted emission values were used for the STOPS forecasting. Section 4.1 in the revised manuscript contains more detailed description for the method we used for STOPS forecasting with GOCI-derived AOD.

15. P6, line 30-35, what does the windblown dust module do in CMAQ? Any references for other studies using it? Figure 2 comparison shows almost no difference in PM predictions from simulations of standard CMAQ and CMAQ with dust module, even during the period of the dust event. If you lower the C2 threshold in the dust module, will the CMAQ be able to simulate the dust event?

We provided a brief description of the CMAQ in-line windblown dust module and a reference for it in section 3.1 in the revised manuscript.

<Section 3.1 in the revised manuscript>

The module calculates the vertical dust emission flux (F) by following formula described by

Fu et al. (2014).

$$F = \sum_{i=1}^{M}\sum_{j=1}^{N} K \times A \times \frac{\rho}{g} \times S_i \times SEP \times u_* \times ({u_*}^2 - {u_{*_{ti,j}}}^2)$$

where i and j represent the type of erodible land and soil, K is the ratio between vertical and horizontal flux, A is the particle supply limitation, ρ *is the air density, g is the gravitational constant,* S_i *is the area of the dust source,* SEP *is the soil erodible potential,* u_* *is the friction velocity, and* $u_{*_{ti,j}}$ *denotes the threshold friction velocity.*

When we used the threshold values suggested by Fu et al. (2014), which are lower than standard ones, the simulated PM_{10} concentrations over China, particularly in areas adjacent to the Gobi Desert and its downwind side increased as demonstrated by Fu et al. (2014). But the increase in Korea was relatively minimal and the result did not show reasonable agreement with observation. In this study, the average value of the simulated two-meter temperature during the period was 274.87 K, which was significantly lower than that founded by Fu et al. (2014) (286.30 K). The low friction velocity values below the threshold came from the cold weather conditions over the East Asia during the simulation period. We concluded that the employment of the in-line windblown dust module in CMAQ simulations did not provide discernible enhancement in PM_{10} concentrations because of lower friction velocity than the threshold in the module. These are the reason why we thought a new modeling frame work for the prediction of Asian dust event.

16. P7, line 4-20, I think it will be more appropriate to have these paragraphs in section 2.3 to describe how the satellite AOD used for CMAQ evaluations. Then, section 3.2 can focus on presenting the comparison and discussing the underestimation during the dust period.

As suggested the reviewer, we moved the paragraphs in section 3.2 to section 2.3 in the revised manuscript, so section 3.2 can focus on presenting the comparison and discussing the underestimation during the dust period.

17. P8, section 4.1, it is out of place but better to be moved to section 2.2.

We moved section 4.1 (in the previous manuscript) to section 2.2 (in the revised manuscript) as the reviewer suggested.

18. P8, line 32, why the STOPS domain does not cover the whole Korean Peninsula? In this case, is the AQMS station at the east coast not included in the domain?

We found a problem with the initial position of the STOPS domain. The location of domain center was not 40° N, 119° E, but 40° N, 121° E, so we corrected relevant parts in the revised manuscript (Figure 2 and 10).

19. P9, section 4.2, I cannot get the point of the section. Using half of the page, it repeats findings (CMAQ failed to simulation the dust event and STOPS could produce CMAQ's result with mush less computational time) that have already shown in the previous sections. This section should be re-written to be more concise and informative.

We re-wrote section 4 in the revised manuscript to better explain the method we used for a new PM forecasting using STOPS. We added a figure, which briefly describes entire procedures of the PM forecasting using STOPS with GOCI-derived AOD data.

<Figure S3 in the revised supplementary document>

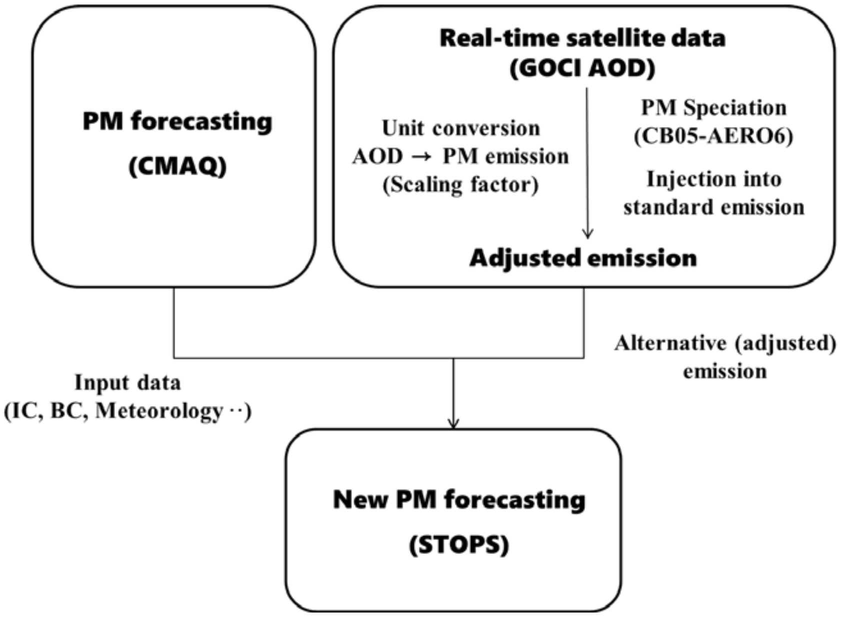


***Figure S3**. Schematic flowchart describing the procedure of the new PM forecasting by STOPS using the real-time AOD data from GOCI.*

20. P9, line 34, I cannot understand how can you add extra amount of PM directly to CMAQ without some kind of data assimilation technique?

Please see our response for question 14.

21. P10, Rather than improving the dust module in CMAQ, using satellite AOD to take into account the extra emission due to the dust event is one reasonable way to improve PM10 prediction for this study. But why the STOPS model is a tool for “a more accurate prediction” (as highlighted in the title)? STOPS is more efficient computationally than running the full CMAQ model? The improvement shown in STOPS results is due to the use of adjusted emission estimated according to the satellite data. By using the same adjust emission, can the CMAQ also produce better PM10 prediction compared to the standard CMAQ?

As the reviewer addressed, the significant improvement in the simulated PM_{10} was contributed by constrained PM concentrations based on GOCI AOD. Even though we used CMAQ instead of STOPS, it would produce the similar results as in STOPS. However, this study assumes a real forecasting situation. In the case of the massive dust transport is captured by satellite measurement, the current forecasting results should be

replaced in a very short time period before the dust storm reaches the receptor regions (Korea in this study). A new forecasting using CMAQ with GOCI AOD cannot be done within a few minutes. Thus, the computational efficiency of STOPS is the most important benefit, which allows the near real-time update of PM forecasting results.

As the reviewer suggested, we revised a couple of misleading sentences throughout the manuscript by saying that STOPS itself does not improve any air quality prediction, but help for "quicker" forecasting.

22. P10, line 32, what is PMT?

The PMT is the same as $PMT_{i,j}$, the estimated emission rate of total PM in each grid cell. We changed PMT to $PMT_{i,j}$ for the better understanding from readers.

23. P11, line 8-16, the text talks about the CMAQ .vs. STOPS simulations but the figure is in CMAQ domains. And the caption indicates both are CMAQ simulations. Please clarify and use consistent names.

We changed Figure 8 in the revised manuscript to show PM_{10} concentrations inside the STOPS domain, and revised its caption for clear description.

<Figure 8 in the revised manuscript>

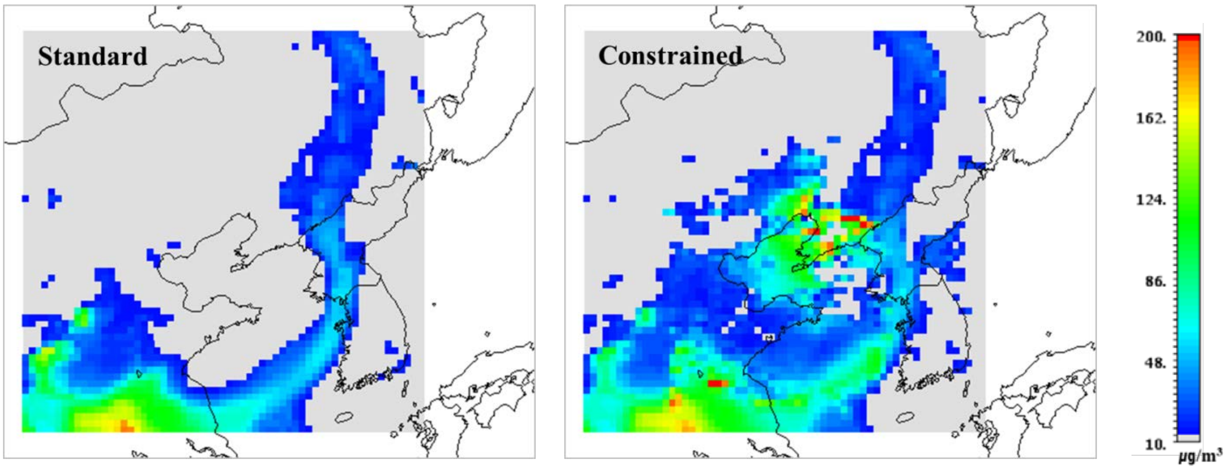


Figure 8. *Difference of the simulated PM_{10} concentrations ($\mu g\ m^{-3}$) between the standard CMAQ run (left) and STOPS forecasting run with alternative emission estimated according to*

GOCI-derived AOD (right) inside the STOPS domain at 12:00 LST on 22 February in 2015.

24. P11, line 8, re-phase it to "Figure 7 shows the comparison of the PM10 concentration from CMAQ simulations using standard and adjusted emission".

P11, line 33-37, I do not know what the "updated" is referring to. Use just "STOPS simulation" instead of "updated STOPS simulation"

P12, line 6-7, re-phase to "the impact of the alternative emissions on the PM10 prediction highly depends on the durations of emission release and the impact was gone after the release ended."

P12, line 17, ': : :AOD data contained missing data due to the cloud cover over the C3 study area : : :"

P13, line 28-29, re-phase to "With reasonable meteorological input, the under-prediction of PM10 concentration was mainly due to the inaccurate estimation of dust emission during this period used in CMAQ."

Thanks. We revised the sentences as the reviewer suggested.

25. Figure 2, the CMAQ_dust simulation should be explained in the text and please briefly describe what is the dust module in CMAQ.

We provided a brief description of the CMAQ in-line windblown dust module and a citation for it in section 3.1 in the revised manuscript. Please see our response for question 15.

26. Figure 7, caption: ": : :alternative emission estimated according to the GOCIderived AOD."

As the reviewer suggested, we corrected the caption for Figure 8 in the revised manuscript.

Computationally efficient air quality forecasting tool: implementation of STOPS model into CMAQ v5.0.2 for a prediction of Asian dust

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Abstract. This study suggests a new modeling framework using a hybrid Lagrangian-Eulerian based modeling tool (the Screening Trajectory Ozone Prediction System, STOPS) for a prediction of an Asian dust event in Korea. The new version of STOPS (v1.5) has been implemented into the Community Multi-scale Air Quality (CMAQ) model version 5.0.2. The STOPS modeling system is a moving nest (Lagrangian approach) between the source and the receptor inside the host Eulerian CMAQ model. The proposed model generates simulation results that are relatively consistent with those of CMAQ but within a comparatively shorter computational time period. We find that standard CMAQ generally underestimates PM_{10} concentrations during the simulation period (February 2015) and fails to capture PM_{10} peaks during Asian dust events (22-24 February, 2015), The underestimated PM_{10} concentration is very likely due to missing dust emissions in CMAQ rather than incorrectly simulated meteorology as the model meteorology agrees well with the observations. To improve the underestimated PM_{10} results from CMAQ, we used the STOPS model with constrained PM concentrations based on aerosol optical depth (AOD) data from Geostationary Ocean Color Imager (GOCI), reflecting real-time initial and boundary conditions of dust particles near the Korean Peninsula. The simulated PM_{10} from the STOPS simulations were improved significantly and closely matched to surface observations. With additional verification of the capabilities of the methodology on emission estimations and more STOPS simulations for various time periods, STOPS model could prove to be a useful tool not just for the

predictions of Asian dust but also for other unexpected events such as wildfires and upset emissions events.

1 Introduction

Particulate matter (PM) is one of the key air pollutants in the lower atmosphere. Numerous studies have reported its adverse effects on human health and the environment (Park et al., 2005; Heo et al., 2009; Jeon et al., 2015). Extreme levels of PM and the frequent occurrence of high PM events in the East Asia region have become a major social issue, particularly in South Korea (Korea, hereafter). This is because the region is geographically downwind from China and several desert areas, which are the source of significant emissions. Dust emissions from Mongolia and the Gobi Desert (Chun et al., 2001; Kim, 2008; Heo et al., 2009) cause extraordinarily severe yellow sand storms that often cover the entire sky over Korea during the spring and late winter. These result in the reduced visibility (Chun et al., 2001) and increased mortality due to cardiovascular and respiratory diseases (Kwon et al., 2002), and their adverse effects are more evident in cities closer to source regions of the Asian dust (Kashima et al., 2016).

In response to the problems resulting from Asian dust, the Ministry of Environment of Korea has undertaken $PM_{2.5}$ and PM_{10} forecasting since 2015 to prevent possible harm caused by high PM concentrations; but the forecasting, however, sometimes fails to capture high-level PM events. Accurate PM forecasting is challenging because of the complicated physical and chemical properties of PM and uncertainties in meteorology and emissions (Gelencser et al., 2007; Kim et al., 2008; Tie et al., 2009).

A number of modeling studies have shown the important role of meteorology in PM (Pai et al., 2000; Otte, 2008a; Otte, 2008b), and some have suggested a variety of optimization techniques for enhancing the accuracy of meteorology (Ngan et al., 2012; Lee et al., 2011b; Choi et al., 2012; Jeon et al., 2014; Jeon et al., 2015; Li et al., 2016). Additionally, accurate and updated emission inventories are essential to more accurate PM forecasting. Several studies have used anthropogenic emissions inventories for the Asia domain, such as the International Chemical Transport Experiment - Phase B (INTEX-B) emissions inventory in 2006 and a mosaic Asian anthropogenic emissions inventory in 2010 (MIX) for reliable model performance (Zhang et al., 2009; Zhao et al., 2012; Li et al., 2015). However, the use of the optimized meteorology and the most recent emissions inventory as input data for PM simulations can provide accurate forecasting results for only “normal” time periods, not “upset” events such as Asian dust. This problem is further exacerbated because of the high uncertainty in dust emissions.

To address this issue, the intent of this study is to introduce a modeling tool for PM simulation that can be used in conjunction with the Community Multi-scale Air Quality (CMAQ) model (Byun and Schere, 2006) to more accurately predict PM concentrations, using an Asian dust-storm event as a case-study. We apply a hybrid Eulerian-Lagrangian model, the Screening Trajectory Ozone Prediction System (STOPS), to simulate PM in the East Asia region. The model set-up includes a moving nest domain between the source and the receptor inside the host CMAQ structure. STOPS provides simulation results similar to those of CMAQ, but it does so much faster than the full CMAQ modeling system. Additional details of the original version of STOPS (v1.0) and its effectiveness for regional air quality simulations are explained by Czader et al. (2015). However, since STOPS v1.0 was based on CMAQ v4.4, it is incompatible for recent PM simulations due to outdated modules and chemical mechanisms. Hence, we have implemented a new version of STOPS (v1.5) into CMAQ v5.0.2, which can be utilized with recent emissions inventories, improved chemical mechanisms and useful analyzing tools for the better simulation of Asian dust events.

The primary purpose of this study is to characterize underestimated PM concentrations by standard CMAQ and to determine the primary reason why CMAQ does not accurately capture PM peaks, during the Asian dust events. We introduce a new modeling framework using STOPS as an alternative to full CMAQ modeling for the prediction of severe dust storms over the Korean Peninsula. We utilize STOPS for PM modeling and constrain PM concentrations using real-time satellite data from the Geostationary Ocean Color Imager (GOCI) sensor that allow STOPS to take into account the mostly updated input data (e.g., initial and boundary conditions and emission estimates) inside the modeling domain. We conduct several STOPS simulations using constrained PM concentrations and compare the results to corresponding surface observations to investigate whether the constrained PM concentrations produce accurate PM simulations. We ultimately conclude by proposing the STOPS forecasting/modeling system as an effective tool for capturing severe dust events over East Asia, particularly in Korea.

2 Methodology

2.1 STOPS

STOPS is a hybrid Eulerian-Lagrangian-based modeling tool derived from the CMAQ model. As shown in Fig. 1, a small sub-domain of STOPS was configured inside the CMAQ domain and it moves along with the mean wind from CMAQ. Since STOPS inherits meteorological fields and initial and boundary conditions from a “host” CMAQ simulation, the movement of the STOPS domain is limited to the domain of the host CMAQ simulation. STOPS has the same vertical structure and simulates the same physical and chemical processes as CMAQ, except for the calculation of advection fluxes. CMAQ uses horizontal wind velocity (u and v) from WRF to calculate horizontal advection fluxes, but STOPS calculates the difference between a cell horizontal wind velocity and the mean horizontal velocity in STOPS domain (Czader et al., 2015), so it can consider the moving speed and direction of STOPS domain for the calculation of advection fluxes. Since the STOPS domain moves over time, the horizontal velocity from WRF should be adjusted based on the movement of STOPS domain. The movement of the STOPS domain is determined by the layer-averaged horizontal wind in the center column from the bottom layer up to the top of planetary boundary layer (PBL), weighted by the layer thickness. The averages of the u and v components are calculated by the following equations (Eq. (1)-(2)):

$$\bar{u} = \frac{1}{\sum_{L=1}^{PBLH} \Delta\sigma_F(L)} \sum_{L=1}^{PBLH} u_L \cdot \Delta\sigma_F(L) \qquad (1)$$

$$\bar{v} = \frac{1}{\sum_{L=1}^{PBLH} \Delta\sigma_F(L)} \sum_{L=1}^{PBLH} v_L \cdot \Delta\sigma_F(L) \qquad (2)$$

where $\sigma_F = 1 - \sigma$ and σ is the scaled air pressure in a sigma coordinate system (dimensionless) defined as:

$$\sigma = \frac{(p - p_t)}{(p_s - p_t)} \qquad (3)$$

where p, p_t and p_s denote air pressure at the current level and the top and surface levels of the model, respectively. Czader et al. (2015) present more details on the model and its applications. The first version of STOPS (v1.0) was based on CMAQ v4.4 (Czader et al., 2015), but in this study, it has been updated to v1.5, and implemented in CMAQ v5.0.2.

2.2. Modeling system

In this study, we configured the CMAQ (v5.0.2) model with a domain in grid resolution of 27 km (174 × 128) covering the northeastern part of Asia (Fig. 2), and with 27 vertical layers extending from the surface to 100 hPa. This CMAQ domain, which is slightly larger than standard domain for East Asia study suggested by the Clean Air Policy Modeling System (CAPMOS) (http://capmos.nier.go.kr/index.jsp) of the National Institute of Environmental Research (NIER) in Korea, covers more areas of Gobi Desert which is a major source of Asian dust.

Anthropogenic emissions for the CMAQ domain were obtained from the MIX emissions inventory in 2010 (Li et al., 2015). This inventory contains gridded (0.25° × 0.25°) emissions information for black carbon (BC), carbon monoxide (CO), carbon dioxide (CO_2), nitrogen oxides (NO_x), ammonia (NH_3), organic carbon (OC), fine and coarse particulate matter ($PM_{2.5}$ and PM_{10}), sulfur dioxide (SO_2) and non-methane volatile organic compounds (NMVOC). To acquire high-resolution (1 km × 1 km) anthropogenic emissions in Korea, this study also used the Clean Air Policy

Support System (CAPSS) emissions inventory in 2011 of NIER (Lee et al., 2011a). The CAPSS inventory contains area, line, and point sources of CO, NH_3, NO_x, sulfur oxides (SO_x), total suspended particles (TSP), PM_{10}, and VOC. The emissions for the CMAQ simulations were prepared by the Sparse Matrix Operator Kernel Emissions (SMOKE) model (Houyoux et al., 2000). The Carbon Bond chemical mechanism (CB05) (Yarwood et al., 2005) and the AERO6 aerosol module (Nolte et al., 2015) were used for gas-phase and aerosol chemical mechanisms respectively, and initial and boundary conditions were obtained from the standard CMAQ profile.

We simulated meteorological fields using the Weather Research and Forecast (WRF, v3.7) model (Skamarock et al., 2008) and used the 1°× 1° Final Operational Global Analysis (FNL) data of the National Centers for Environmental Prediction (NCEP) to determine the initial and boundary conditions. To improve the accuracy of meteorological fields, we adopted the optimized grid analysis nudging options suggested by Jeon et al. (2015) for the East Asia simulations.

The WRF-CMAQ simulations were conducted for the period of 21 January to 28 February in 2015, which included the first ten days for spin-up. Evaluations applied to the month of February and the three-day Asian dust event occurred during 22-24 February in 2015. During the event days, massive dust over the GOBI desert and Mongolia region was transported to the Korean Peninsula. This happened due to the southeastward wind resulting from high pressure over the Mongolia region and low pressure over the northeastern part of China (Fig. S1 in the supplementary document). The detailed options used for WRF and CMAQ simulations are listed in Table S1 and S2 in the supplementary document.

The configuration of the CMAQ sub-domain for the STOPS simulation consists of 61 × 61 horizontal grid cells that covers a portion of the Korean Peninsula and the Yellow Sea, and its initial position was near the northern part of the Yellow Sea (40° N, 121° E) (Fig. 2), the transporting pathway of Asian dust. The simulated PM_{10} concentrations of standard STOPS during Asian dust events (22-24 February, 2015) closely agreed with those of CMAQ (Fig. S2 in the supplementary document). The correlation coefficients (R) for each day were 0.94, 0.96, and 0.97, indicating that the results from STOPS and CMAQ are significantly correlated. This reasonable consistency of STOPS and CMAQ results justifies the use of STOPS instead of CMAQ, in this study.

2.3 In-situ and satellite measurements

This study used surface observational data from the Air Quality Monitoring Station (AQMS) network operated by NIER. The network measures real-time air pollutant concentrations and provides hourly concentrations for CO, NO_2, O_3, $PM_{2.5}$, PM_{10}, and SO_2. We gathered the measured $PM_{2.5}$ and PM_{10} data in 2015 from the AQMS network and while meteorological data were obtained from the Automatic

Weather System (AWS) network, operated by Korea Meteorological Administration (KMA). The following statistical parameters were used for the evaluation of the performance of WRF and CMAQ simulations: Index Of Agreement (IOA), Mean Bias Error (MBE) and Root Mean Square Error (RMSE). These are defined as:

$$\mathrm{IOA} = 1 - \frac{\sum_{i=1}^{N}(P_i - O_i)^2}{\sum_{i=1}^{N}(|P_i - \bar{P}| + |O_i - \bar{O}|)^2}$$

$$\mathrm{MBE} = \frac{\sum_{i=1}^{N}(P_i - O_i)}{N}$$

$$\mathrm{RMSE} = \sqrt{\frac{\sum_{i=1}^{N}(P_i - O_i)^2}{N}}$$

where N is number of data points and P_i and O_i denote CMAQ-simulated and observed concentrations, respectively.

We also employed the aerosol optical depth (AOD), measured by a GOCI sensor from the geostationary orbit onboard the Communication Ocean and Meteorological Satellite (COMS). The GOCI level 1B (L1B) data provide hourly daylight spectral images (09:30-16:30 LST, 8 times a day) for East Asia. The spatial coverage extends to 2500 km × 2500 km centered at 36° N, 130° E (Lee et al., 2010; Choi et al., 2016). The AOD at 550 nm with a 6 km resolution were obtained from GOCI L1B data, using a retrieval algorithm introduced by Choi et al. (2016). The GOCI-derived AOD data were used for constraining of PM concentrations and the model evaluation. For the evaluation of CMAQ simulated PM_{10}, we converted the concentration units in CMAQ to AOD for a fair comparison of the results with GOCI. The aerosol properties from the CMAQ simulation (CMAQ-derived AOD) were obtained by the following equations (Eq. (4)-(6)), which were introduced by Roy et al. (2007) and have successfully been tested in East Asia (Song et al., 2008; Park et al., 2011):

$$AOD_{CMAQ} = \sum_{i=1}^{N}(\sigma_{sp} + \sigma_{ap})_i \Delta Z_i \tag{4}$$

$$\sigma_{sp} = (0.003)f_t(RH)[NH_4{}^+ + SO_4{}^- + NO_3{}^-] + (0.004)[OM] + (0.001)[FS] + (0.0006)[CM] \quad (5)$$

$$\sigma_{ap} = (0.01)[LAC] \quad (6)$$

where i is the vertical layer number, ΔZ is the layer thickness, and the brackets indicate mass concentrations in mg m^{-3} units. The OM, FS, CM and LAC denote mass concentrations of organic species, fine soil, coarse particles, and light-absorbing carbon, respectively. The specific scattering coefficients in the equations (i.e., 0.003, 0.004, 0.001, 0.0006, and 0.001) are represented in units of m^2 mg^{-1}. The $f_t(RH)$, calculated by the method described by Song et al. (2008), denotes relative humidity based on the aerosol growth factor.

3 PM$_{10}$ simulation results from standard CMAQ

3.1 Comparison with surface measurement

We simulated PM$_{10}$ concentrations by standard CMAQ and compared them with surface observational data obtained from the AQMS network of NIER in Korea. For this comparison, we selected 20 AQMS sites, evenly distributed in Korea (Fig. 2), and calculated mean PM$_{10}$ concentrations at all of the sites. We do not present the results for PM$_{2.5}$ because the simulated PM$_{2.5}$ exhibited almost same temporal variation and lower concentrations to those of PM$_{10}$. In addition, the coarse particles comprise a major portion of the total PM during the Asian dust period, as described by Chun et al. (2001). From the comparison shown in Fig. 3, the concentration of CMAQ-simulated PM$_{10}$ was slightly underestimated, but its temporal variation showed reasonably close agreement with observation except for the Asian dust episode (22-24 February, 2015). The CMAQ failed to capture the high peaks of PM$_{10}$ in the episode caused by the transport of massive dust from the Gobi Desert and Mongolia region.

As shown in Table 2, the performance of CMAQ simulation for the entire period (February 2015) was poor. For example, the high value of RMSE (78.03 μg m^{-3}), low value of IOA (0.36) and negative value of MBE (-39.94 μg m^{-3}) indicate that CMAQ underestimated PM$_{10}$, and its temporal variation did not agree well with the observations. The calculated statistics for the period excluding the Asian dust episodes was much better than those for the entire period, as indicated in Table 2. The large differences in these findings clearly reveal that the performance of CMAQ is relatively accurate for the regular simulation period, but it is not for the Asian dust period. As shown in Fig. 4 and Table S3, meteorological fields such as temperature and wind speed in the receptor regions (Korea) showed close agreement with observations, even during the Asian dust period. It suggests that the underestimated PM$_{10}$ concentration was likely due to the uncertainty in meteorology in the source regions (China and

Mongolia), and/or faulty estimation of dust emissions for the CMAQ simulation. We attributed the main reason for the PM_{10} underestimation to poorly estimated dust emission because CMAQ showed poor performance only during the Asian dust period.

To enhance the performance of CMAQ for PM_{10} simulations during the Asian dust period, we employed the in-line windblown dust module in CMAQ v5.0.2. The module calculates the vertical dust emission flux (F) by following formula described by Fu et al. (2014).

$$\mathrm{F} = \sum_{i=1}^{M} \sum_{j=1}^{N} K \times A \times \frac{\rho}{\mathrm{g}} \times S_i \times SEP \times u_* \times ({u_*}^2 - {u_{*ti,j}}^2) \quad (7)$$

where i and j represent the type of erodible land and soil, K is the ratio between vertical and horizontal flux, A is the particle supply limitation, ρ is the air density, g is the gravitational constant, S_i is the area of the dust source, SEP is the soil erodible potential, u_* is the friction velocity, and $u_{*ti,j}$ denotes the threshold friction velocity.

Interestingly, the employment of the in-line windblown dust module in CMAQ simulations did not provide discernible enhancement in PM_{10} concentrations (Table 2) because of lower friction velocity than the threshold in the module during the simulation period (February 2015) (Table S4 in the supplementary document). Several studies have reported that the threshold friction velocity plays a key role in the calculation of dust emission flux because the threshold can determine the probability of the lifting of dust particles (Choi et al., 2008; Fu et al., 2014). This research also implies that more studies that enhance the capability of dust modules during the winter period should be performed.

3.2. Comparison with satellite-based observation

Figure 5 presents a comparison of time-averaged AOD derived from GOCI and CMAQ. For an unbiased comparison of AOD, we removed grid cells from GOCI data consisting of fewer than 15 pixels (i.e., bad pixels) because of cloud contamination; we also did not include the corresponding grid cells in CMAQ for our comparison. The GOCI-derived AOD shows several blank areas in the northern part of the Korean Peninsula, near the northeastern region of China, and in most regions of Japan because of the significantly high fraction of clouds over these areas. The horizontal features of the CMAQ-derived AOD were similar to those of the GOCI-derived AOD, but CMAQ overestimated the AOD near the southeastern part of China. On the other hand, compared to the GOCI-derived AOD, the CMAQ underestimated the AOD over the Yellow Sea and Korea. As mentioned in Sect. 3.1, CMAQ underestimated surface PM_{10} concentrations in Korea. The CMAQ-derived AOD in Korea was also underestimated compared to GOCI-derived AOD, consistent with the surface measurements. These comparisons using the satellite and surface measurements indicated that the CMAQ was unable to capture the high levels of PM in Korea during the simulation period in this study (February 2015).

Hence, the discrepancy between CMAQ- and GOCI-derived AOD is likely due to uncertainty in emissions of PM precursors such SO_2, NO_x and NH_3 (Jeon et al., 2015) and meteorology over source regions as discussed in Sect 3.1.

Compared to the GOCI-derived AOD, the CMAQ-derived AOD near the northern regions of the Korean Peninsula was underestimated. This underestimation may have resulted from the failure of CMAQ to simulate the Asian dust emissions and their transport to the Korean Peninsula on 22-24 February in 2015. The CMAQ-derived AOD was underestimated primarily in the moving pathway of the Asian dust (i.e., between the Gobi Desert (source area) and Korean Peninsula (receptor area)). As addressed in Sect. 3.1, the in-line windblown dust module in CMAQ failed to accurately estimate the dust emissions during the Asian dust period and it caused the model to underestimate AOD near the northern regions of the Korean Peninsula.

To further investigate the issue of underestimation of CMAQ during the period of Asian dust (22-24 February, 2015), we compared the GOCI- and CMAQ-derived AODs on each event day. Unfortunately, the comparison was available only on 22 February since the GOCI-derived AOD included a significantly high number of blank pixels on the other event days because of the high fraction of cloud cover. Figure 6 shows GOCI- and CMAQ-derived daily mean (09:30-16:30 LST) AODs on 22 February. The GOCI-derived AOD clearly showed massive dust near the northwestern regions of the Korean Peninsula and the eastern part of China and densely distributed dust particles over the Yellow Sea that were transported from the Gobi Desert. In contrast, CMAQ did not reproduce the high dust concentrations near the Korean Peninsula because of the failure in the estimation of dust emissions.

We concluded that CMAQ clearly underestimated PM_{10} concentrations during the simulation period and failed to capture peaks during the Asian dust period starting on 22 February. Thus, we attempted to use STOPS for capturing the dust enhanced PM_{10} in Korea (receptor region). We used the dust storm data temporarily detected by satellite measurements between the source and receptor regions as an input for the STOPS modeling. The following sections describe the details how STOPS was used for PM_{10} forecasting.

4 Application of STOPS for PM_{10} forecasting

Assuming the CMAQ PM_{10} simulation results in this study were used for forecasting purposes, the severe dust events starting on 22 February in 2015 could not be predicted. Thus, to accurately forecast the transport of massive dust storm, we must take into account the most recent and accurate initial and boundary conditions and emissions. Figure 7 shows the GOCI-derived AOD on 21-22 February, when a dust storm was approaching Korea (receptor region). The massive dust storm was not evident from the GOCI-derived AOD on 21 February, but a center of the dust storm in the northwestern region of the Korean Peninsula was first seen at 10:30 LST on 22 February. Upon observation of the massive dust from the GOCI-derived AOD at 10:30 LST on 22 February, a new PM_{10} forecasting using STOPS with real-time AOD data can be performed in a short time (i.e. a few minutes) and the current forecasting results can be replaced by the results from the STOPS. For the new PM_{10} forecasting using STOPS, we intended to use the GOCI-derived AOD as a new initial condition for PM_{10} species. However, the approach does not fully consider continuous transport of dust from the source regions because the impact of the changed initial condition on the STOPS results would be diminished within a few hours. Thus, we used the GOCI-derived AOD as PM emissions for the STOPS forecasting to make the best use of the AOD data.

4.1 Satellite-adjusted PM concentrations

For the new PM_{10} forecasting using STOPS, we first attempted to convert the GOCI-derived AOD to PM concentrations and directly add them to the simulated PM concentrations by STOPS. However, the sudden and rapid changes in PM concentration made the STOPS simulation unstable and they sometimes caused unexpected termination of STOPS runs due to overflow error. To resolve this problem, we regarded the GOCI-derived AOD as PM emissions and indirectly constrained the original PM concentrations by using the alternative emissions. In short, the GOCI-derived AOD was converted to emissions and used for the STOPS forecasting. We should note that the alternative emissions are not real, but the enhanced dust concentrations which are taking the form of emissions. We concluded this methodology could be an effective way to reflect the satellite measured AOD to CMAQ simulation without possible computational error.

As indicated in Fig. 7, the massive dust storm was first captured by the GOCI-derived AOD at 10:30 LST on 22 February in 2015, so we adjusted the standard emissions at a corresponding time based on the GOCI-derived AOD and used them for the STOPS forecasting. We should note that the AOD and the emissions rate are expressed in different units; the AOD is a unitless value, while the emission rate is expressed in units of grams per second (particles) or moles per second (gas-phase species); therefore, we employed a scaling factor to convert the AOD to the emissions rate. To find a reasonable scaling factor, we re-gridded the domain of the GOCI-derived AOD data so that it corresponded to the CMAQ domain and compared the AOD in each grid cell with corresponding emission rates of total PM in the MIX inventory (e.g., PM_{10}). We used only the grid cells with valid AODs (no missing values) and emission rate (> 0) for the comparison and then calculated the average ratio of the AOD to emissions rates. The calculated ratio was 1,884.49 $g\ s^{-1}$ for this case, indicating that the emissions rate of total PM inside the modeling domain was 1,884.49 times larger than the GOCI-derived AOD. It should be noted that the ratio cannot generally explain the relationship between AOD and emissions. Because the relationship is valid for only a particular domain (Fig. 2) and time (10:30 LST on 22 February, 2015), the ratio for each case should be recalculated.

For the unit conversion from the AOD to the emissions rate of total PM, we used the estimated ratio as a scaling factor and calculated the total PM emissions by the following equation (Eq. (8)):

$$PMT_{i,j} = AOD_{i,j} \times \mathrm{SF} \tag{8}$$

where $PMT_{i,j}$ and $AOD_{i,j}$ represent the emission rates of total PM and GOCI-derived AOD in each grid cell, respectively. SF is the calculated scaling factor (1,884.49 g s^{-1}), which indicates the relationship between the AOD and the emissions rate.

For the STOPS simulation, we split the calculated $PMT_{i,j}$ into several specific species, including coarse and fine particles, used for the CB05-AERO6 chemical mechanism. In order to calculate the species distribution, we estimated the mass fractions of each PM species during the Asian dust events based on the findings in Kim et al. (2005) and Stone et al. (2011), which described the composition of measured PM during the Asian dust periods (Table 3). More than half of the $PMT_{i,j}$ was allocated to coarse particles (PMC) because they comprise a major percentage of Asian dust, as reported in several studies (Kim et al., 2003; Lee et al., 2004; Kim et al. 2005; Stone et al., 2011). The speciated PM emissions were injected into standard PM emissions in each grid cell. Based on the findings by Kim et al. (2010), the amounts of the alternative emissions were assumed to be distributed below the altitude of 3 km (1 to 11 vertical layers). The entire procedures of the new PM forecasting by STOPS using GOCI-derived AOD are briefly depicted in Fig. S3.

4.2 Enhanced PM_{10} forecasting using STOPS

We conducted a new PM_{10} forecasting run using STOPS with the constrained PM concentrations (by using alternative emissions) and examined the improvement in its accuracy over that of the standard CMAQ model. The STOPS forecasting covers one-day (24 hours), which began at 11:00 LST on 22 February in 2015 immediately following the massive dust first observed in the GOCI-derived AOD between the source and receptor regions.

Figure 8 shows the comparison of the PM_{10} concentration from CMAQ using standard emissions and STOPS using alternative emissions. The PM_{10} from standard CMAQ exhibited high concentrations over the eastern part of China, central Yellow Sea and northwestern part of the Korean Peninsula. By contrast, the constrained PM_{10} by the alternative emissions (Fig. S4 in the supplementary document) exhibited significantly increased concentration, particularly in the northwestern part of the Korean Peninsula (Fig. 8). The constrained PM_{10} concentration showed similar features as those of the GOCI-derived AOD, shown in Fig. 6-(a), implying that the dense dust attributed by Asian dust were accurately reflected in the STOPS forecasting.

We should note that the duration of the release of alternative emissions strongly affected the simulated $PM_{10.}$ Hence, it plays an important role in the STOPS forecasting, so we conducted four forecasting runs with different release durations (3hr, 6hr, 12hr, and 24hr) as shown in Fig. 9, and compared all of the results with those of standard CMAQ and available PM_{10} surface measurements. Figure 9 exhibits clear differences in the temporal variation of PM_{10} resulting from the impact of the durations. As addressed in Sect. 3.1, the standard CMAQ run failed to capture the drastic increase in PM_{10} concentrations on 22 February in 2015 because of the poor dust emission modeling in CMAQ. However, the STOPS forecasting showed significantly improved PM_{10} results compared to standard CMAQ. The results indicated higher PM_{10} concentrations than those of CMAQ, and they were much closer to observations.

Interestingly, Figure 9 shows that predicted PM_{10} by STOPS with a duration of release of three hours (STOPS_E3) closely agreed with observations during the first three hours. However, the simulated PM_{10} began to decrease immediately after the third hour, and the agreement with observations gradually worsened with time. The results of the other STOPS runs with different durations of release of 6, 12, and 24 hours (STOPS_E6, STOPS_E12 and STOPS_E24, respectively) were almost the same as those of STOPS_E3. In other words, the impact of the alternative emissions on the PM_{10} prediction highly depends on the durations of emission release and the impact was gone after the release ended. STOPS_E24 represented the closest agreement with observations, implying that STOPS_E24 produced the greatest improvement in one-day PM_{10} forecasting because of continuous emissions during the entire forecasting time (24 hours).

Despite its positive performance in one-day PM_{10} forecasting, STOPS_E24 did not perfectly capture the high PM_{10} concentrations during the Asian dust event. In fact, it underestimated the peak of observed PM_{10}, which may have resulted from uncertainty inherent in the methodology using AOD estimation. Direct conversion from the AOD to the alternative emissions rate using a scaling factor is challenging because it has not yet proven reliable by existing studies. Hence, the uncertainty inherent in unit conversion might have contributed to the inaccuracy of the emissions rate. In addition, the GOCI-derived AOD data contained missing data due to the cloud cover over the study area during the event on 22 February, and as a consequence, it did not accurately represent the distribution of transported Asian dust. The most probable reason for the underestimated PM_{10} simulated by STOPS was that the alternative emissions during the first time step (11:00 LST on 22 February, 2015) were subsequently used for all of the time steps without accounting for spatiotemporal variations. Since the horizontal and vertical distributions of the Asian dust changed with time, the alternative emissions in the first time step did not accurately represent the varied dust distribution in the next time step. The uncertainty with regard to the alternative emissions increases with time. The STOPS_E24-predicted PM_{10} concentrations showed close agreement with observations during the first six hours (Fig. 9), but error gradually widened with time. However, as observation in later hours cannot be reflected at the beginning of forecasting, such a problem is inevitable in a forecasting mode. Thus, repeated forecasting for short time periods (e.g., six hours) with the variable alternative emissions could possibly provide more accurate PM_{10} results for the Asian dust events. STOPS would be very useful for repeated PM_{10} forecasting because of its remarkably short simulation time (a few minutes).

To verify the horizontal distribution of PM_{10} resulting from the effect of constrained PM, we compared the simulated surface PM_{10} concentrations from the STOPS forecasting to those from standard CMAQ. Figure 10 shows the horizontal distribution of surface PM_{10} concentrations inside the STOPS domain simulated by standard CMAQ and STOPS_E24, which indicates the most accurate one-day forecasting results of all the STOPS simulations (from Fig. 9). The location of the STOPS domain moved slightly toward a southeasterly direction according to the changed mean wind in the domain. In the first time step (0 hr, 11:00 LST, 22 February), STOPS_E24 showed the same PM_{10} distribution as standard CMAQ because the initial condition for the STOPS simulation was provided by the standard CMAQ. After eight hours, the PM_{10} concentration from STOPS_E24 differed from that of the standard CMAQ owing to the effect of the alternative emissions by the GOCI-derived AOD. After sixteen and twenty four hours, the difference became more pronounced. Results of standard CMAQ did not show a high level of PM_{10}, but those of STOPS_E24 showed a PM_{10} concentration of at least 100 μgm^{-3} near the Korean Peninsula. Specifically, they showed extremely high PM_{10} concentrations of over 1,500 μgm^{-3} in the northwestern part of the Korean Peninsula. Figure 7 (10:30 LST on 22) indicates massive dust over that area from the GOCI-derived AOD consistent with the enhanced PM_{10} concentrations. The massive dust over the region were transported to Korea and led to significantly enhanced levels of PM_{10}. The horizontal distributions of PM_{10} at higher vertical levels up to 3 km showed similar features at the surface layer because the alternative emissions were evenly distributed below that level.

Overall, even with the uncertainties addressed above, the massive dust storm near the Korean Peninsula on an Asian dust day was reasonably reproduced by the STOPS forecasting with using PM emissions constrained by GOCI-derived AOD. These results indicate that the STOPS could possibly be used for new PM_{10} forecasting with real-time constraints of PM concentration and this methodology should enhance the performance of PM_{10} forecasting and modeling.

5 Summary

This study introduced a new modeling framework using a hybrid Eulerian-Lagrangian model (called STOPS) that showed almost the same performance as CMAQ, but used a shorter simulation run-time. STOPS v1.5 has been implemented into CMAQ v5.0.2 for PM_{10} simulations over the East Asia during Asian dust events, and we investigated possibility of using STOPS to enhance the accuracy of PM_{10} forecasting. During the entire simulation period (February 2015), the standard CMAQ underestimated PM_{10} concentrations compared to surface observations and failed to capture the PM_{10} peaks of Asian dust events (22-24 February, 2015). With reasonable meteorological input, the under-prediction of PM_{10} concentration was mainly due to the inaccurate estimation of dust emissions during this period used in CMAQ. We also evaluated the horizontal feature of CMAQ simulated PM_{10} using satellite-observed data (GOCI). The PM_{10} results from the standard CMAQ run were compared to those of the GOCI-derived AOD and the results indicated that the standard CMAQ barely captured the transported dust from the Gobi Desert to the Korean Peninsula during the Asian dust events.

For more accurate PM_{10} prediction, we used the STOPS model and conducted several simulations using constrained PM concentrations (by using alternative emissions) based on the GOCI-derived AOD, which reflected the most recent initial and boundary conditions near the Korean Peninsula. The STOPS simulations showed higher PM_{10} concentrations than the standard CMAQ and indicated clear dependence on the duration of the alternative emission release. The STOPS simulations showed reasonable PM_{10} concentrations close to observational data, but they did not capture the peak during the Asian dust events because of uncertainty in the methodology used for the constraining PM concentrations. The direct conversion from AOD to emissions using a scaling factor was challenging because it has not yet proven reliable by existing studies. In addition, the GOCI-derived AOD data were missing many values because of the high fraction of clouds cover during the event and consequently, it did not accurately reflect the massive dust storm on the Asian dust day.

Overall, STOPS reasonably reproduced the high level of PM_{10} over the Korean Peninsula during the Asian dust event with constrained PM concentrations using satellite measurements. Although STOPS indicated significantly high PM_{10} enhancement for the episode, it still requires improvement before its results can be generalized. Thus, we should direct our study toward additional verification of the methodology regarding unit conversion (e.g. possible nonlinearities) and numerous sensitivity simulations for different cases to determine the optimal duration of the release of the alternative emissions. The results of this study are an ideal starting point for such studies.

The ultimate goal of this study was to suggest an effective tool for successive PM_{10} forecasting and modeling over the East Asia, and the results clearly showed the reliability and various advantages of STOPS modeling. Therefore, because of its reliable performance with remarkably high computation efficiency, the STOPS model could prove to be a highly useful tool for enhancing dust forecasting/modeling performance over East Asia. Further, the benefit of STOPS modeling could be generalized to the forecasting and modeling of unexpected events such as wildfires and upset oil and emissions events.

Code availability

The STOPS v1.5 source code can be obtained by contacting the corresponding author at ychoi6@uh.edu.

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Table 1. Observed PM_{10} and $PM_{2.5}$ concentrations ($\mu g\ m^{-3}$) recorded on each days of an Asian dust event in February 2015. The values are averaged of the 20 AQMS sites shown in Fig. 1. D_Max denotes daily maximum concentrations and D_Mean daily mean concentrations.

	PM_{10}		$PM_{2.5}$	
	D_Max	D_Mean	D_Max	D_Mean
Feb 22	345.47	111.52	28.75	18.85
Feb 23	472.47	341.63	72.67	43.61
Feb 24	175.88	111.86	37.78	23.46

Table 2. Statistical parameters of PM_{10} concentrations at 20 AQMS sites in Korea for the simulations without the dust module (CMAQ), with the in-line dust module (CMAQ_Dust).

	Entire period			Without dust events		
	RMSE	IOA	MBE	RMSE	IOA	MBE
CMAQ	78.03	0.36	-39.94	28.56	0.81	-22.83
CMAQ_Dust	78.03	0.36	-39.94	28.56	0.81	-22.83

Table 3. Specific fractions (%) for the splitting of total PM emission into specific PM species in the CB05-AERO6 chemical mechanism used in this study.

PM Emission Species	Fraction	PM Emission Species	Fraction
PMC (Coarse Particle)	55%	PCA (Calcium)	2%
PMOTHR (Unspeciated $PM_{2.5}$)	25%	PEC (Elemental Carbon)	1%
PSO_4 (Sulfate)	8%	PNA (Sodium)	1%
PNO_3 (Nitrate)	3%	PCL (Chloride)	1%
POC (Organic Carbon)	3%	PK (Potassium)	1%
PNH4 (Ammonium)	2%		

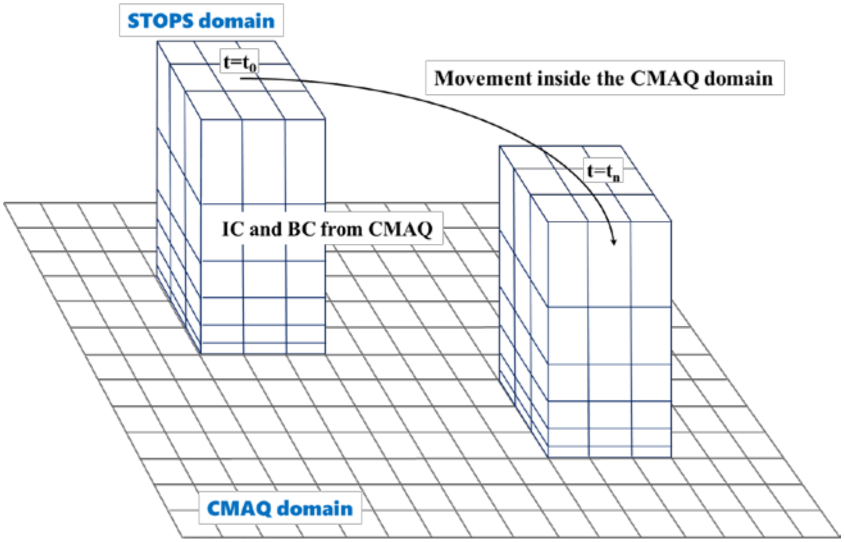

Figure 1. Conceptual diagram showing the basic structure and movement of the STOPS domain inside the CMAQ domain.

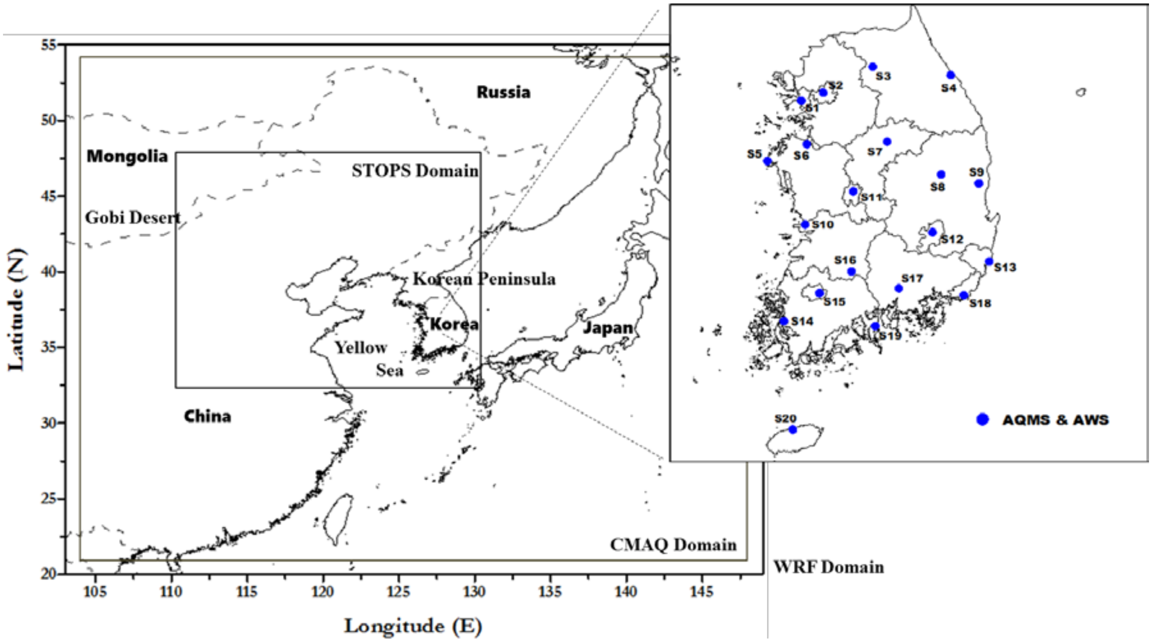

Figure 2. Domains for the WRF, CMAQ and STOPS modeling. The right panel shows the location of the air quality monitoring stations (AQMS) and automatic weather system (AWS) sites used in this study.

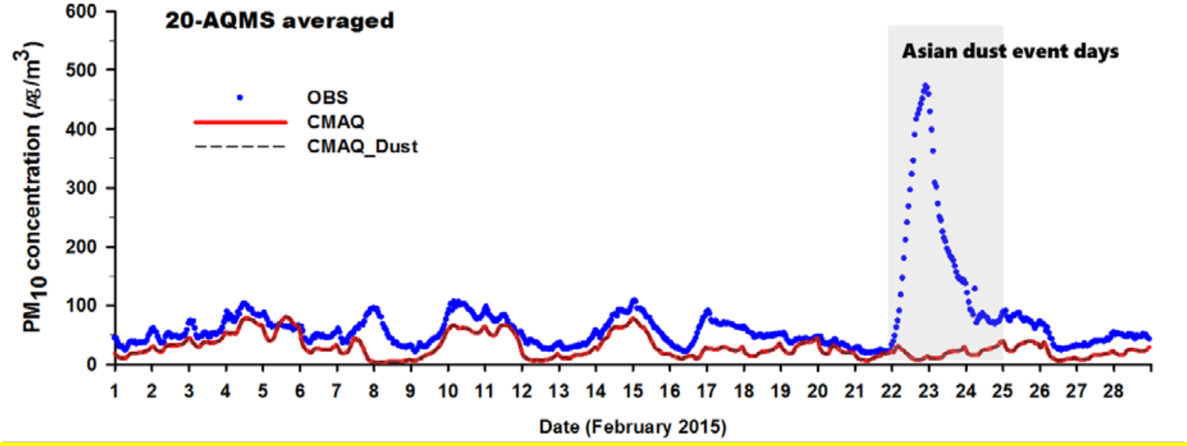

Figure 3. Time series of observed (OBS, blue dots) and simulated (CMAQ: red line, CMAQ_Dust: black dashed line) PM$_{10}$ concentrations in February 2015. The values are averaged values for 20 AQMS sites: CMAQ_Dust is closely coupled with the standard CMAQ modeling results (red line).

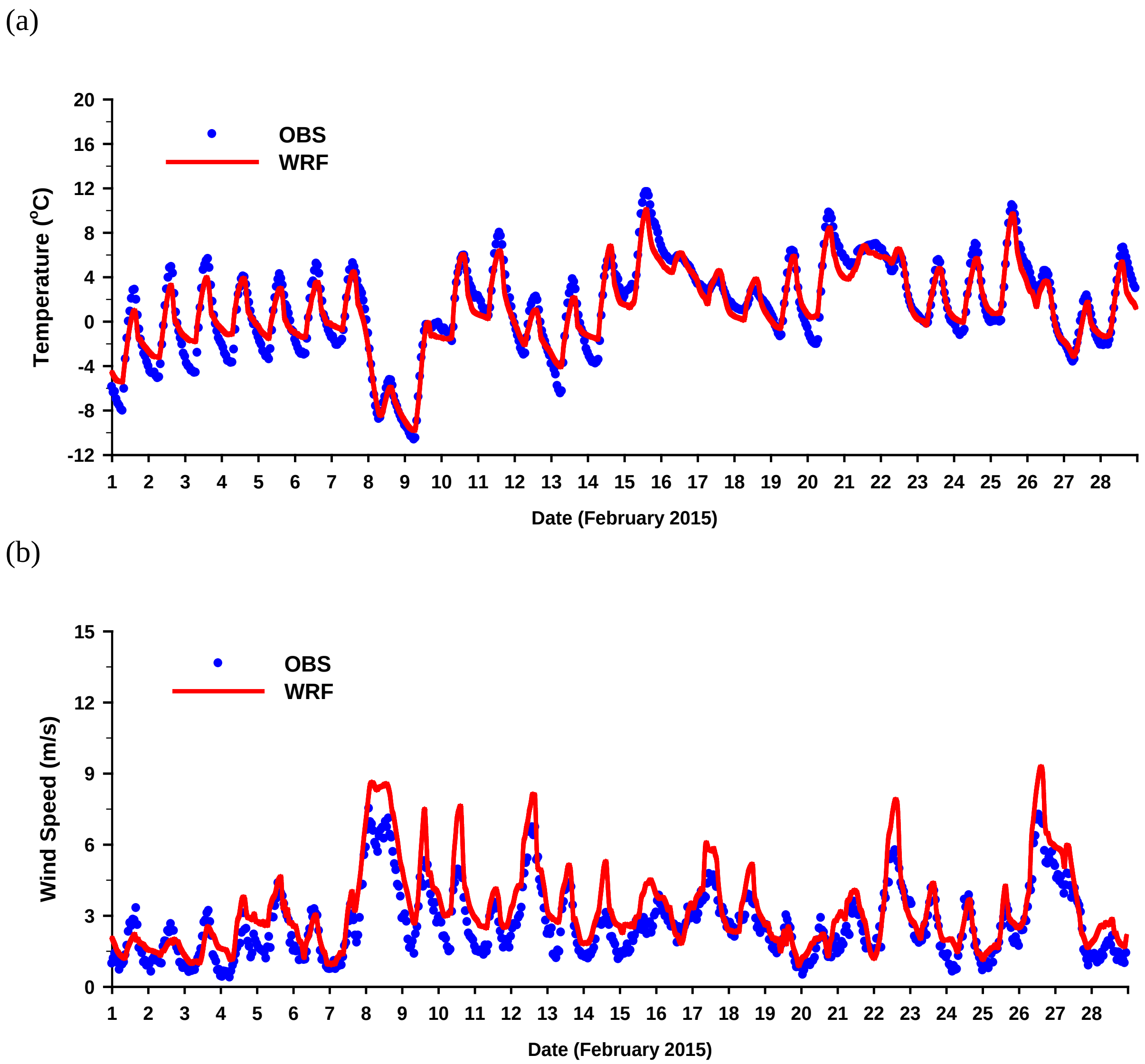

Figure 4. Time series of observed (OBS, blue dots) and WRF simulated (WRF, red line) (a) temperature and (b) wind speed in February 2015. The values are averaged values for 20 AQMS sites.

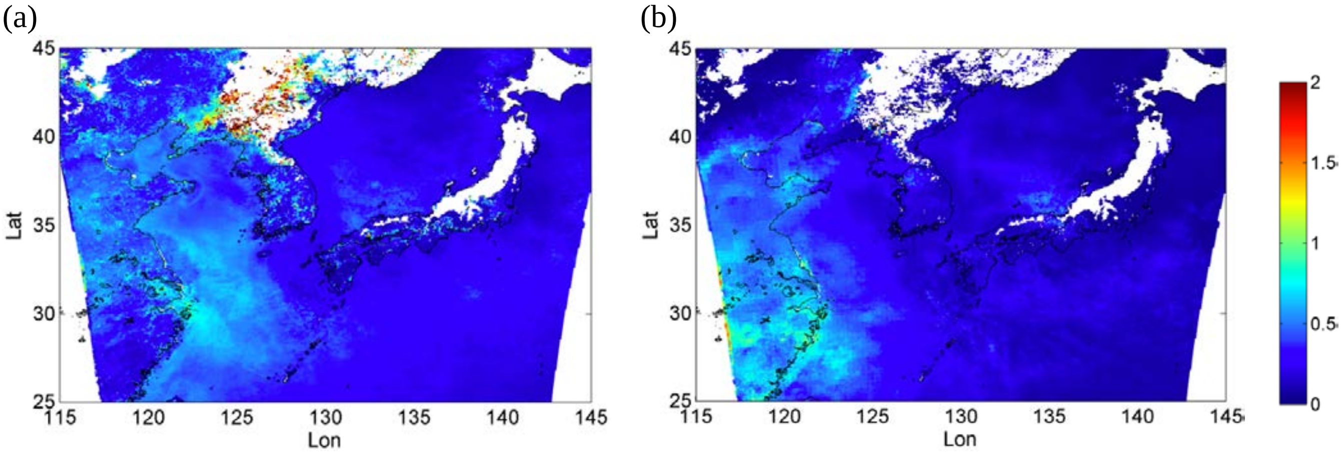

Figure 5. The (a) GOCI- and (b) CMAQ-derived AOD (550 nm) during the entire time period of simulations. The values are averaged for February 2015.

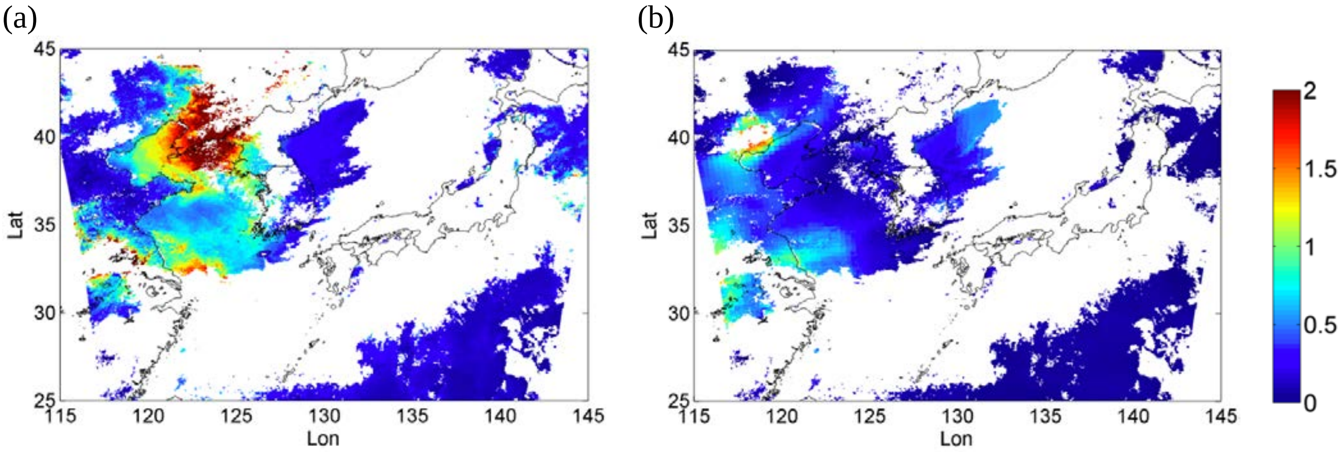

Figure 6. The (a) GOCI- and (b) CMAQ-derived AODs (550 nm) on 22 February. The values are averaged from 09:30 to 16:30 LST.

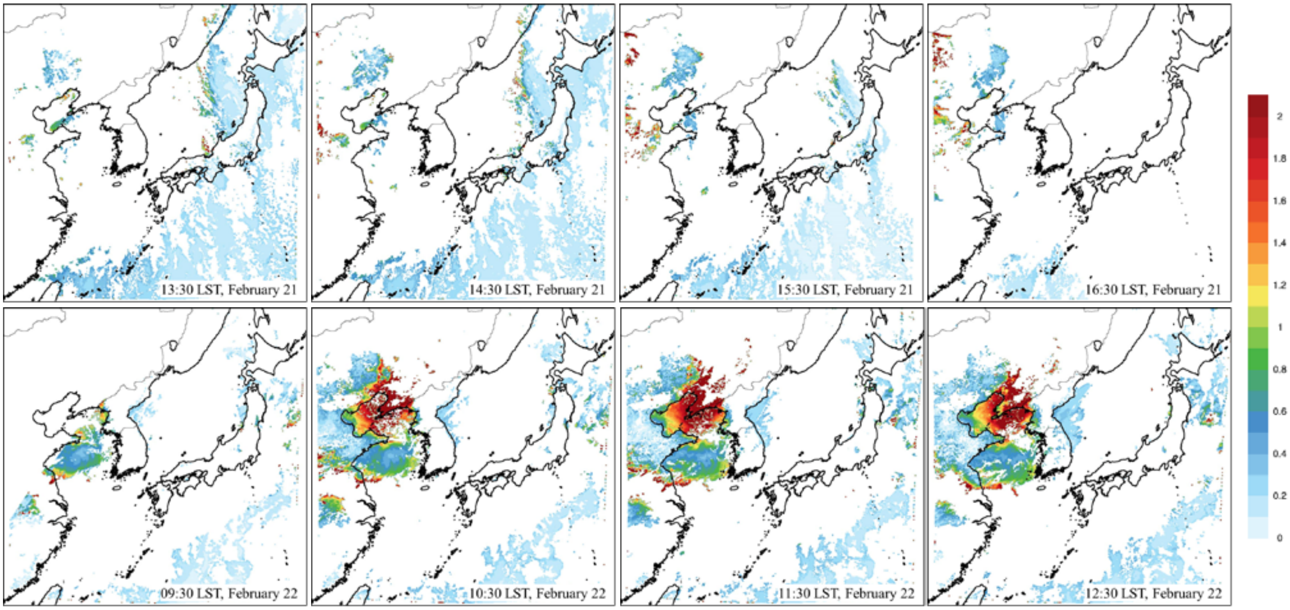

Figure 7. The GOCI-derived AOD (550 nm) from 13:30 LST on 21 February to 12:30 LST on 22 February in 2015. The white-colored areas represent missing pixels.

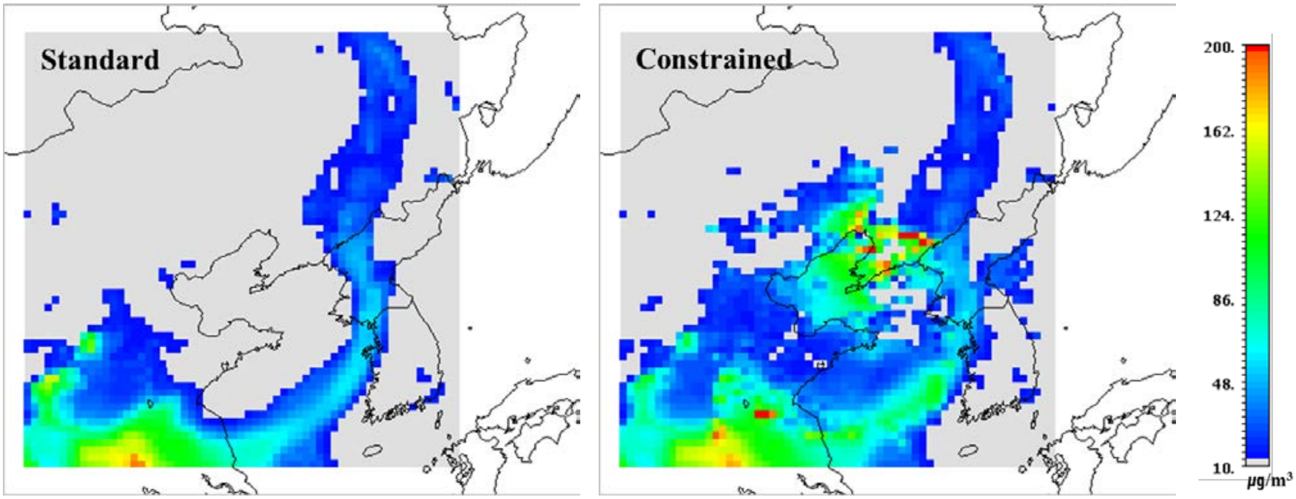

Figure 8. Difference of the simulated PM_{10} concentrations ($\mu g\ m^{-3}$) between the standard CMAQ run (left) and STOPS forecasting run with alternative emission estimated according to GOCI-derived AOD (right) inside the STOPS domain at 12:00 LST on 22 February in 2015.

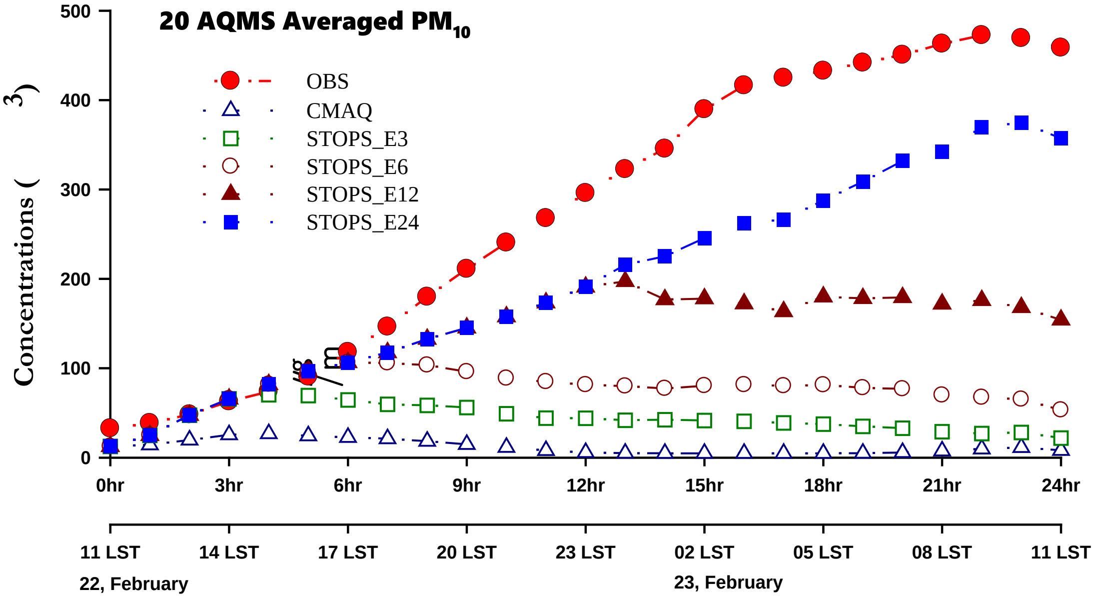

Figure 9. Comparison of observed, CMAQ-simulated and STOPS-simulated PM_{10} concentrations during the 24 hours from 10:00 LST on 22 February in 2015.

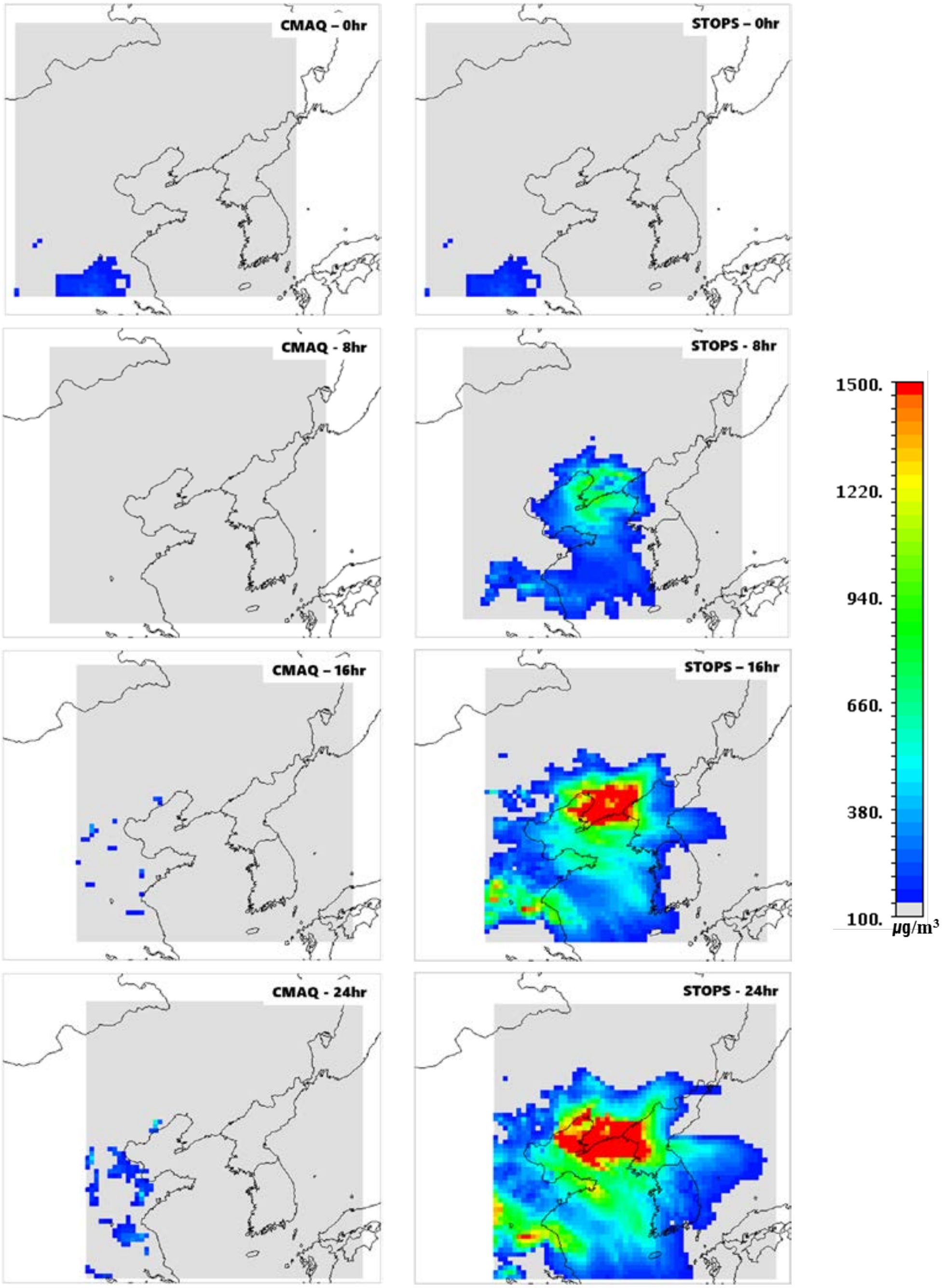

Figure 10. Horizontal distributions of standard CMAQ- and STOPS_E24-simulated surface PM_{10} concentrations inside the STOPS domain. The concentrations were recorded at eight-hour intervals after the beginning of the simulation (11:00 LST on 22 February).

Computationally efficient air quality forecasting tool: implementation of STOPS model into CMAQ v5.0.2 for a prediction of Asian dust

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Supplements

Table S1 and S2 are a supplement for the description of model configuration for WRF and CMAQ simulations in Sect. 2.2 in the manuscript.

Table S3 is a supplement for the description of WRF evaluation results during the time period of simulation at 20 observational sites in Sect. 3.1 in the manuscript.

Table S4 is a supplement for the description of difference between the simulated friction velocity and threshold values in the in-line dust module in CMAQ in Sect. 3.1 in the manuscript.

Figure S1 is a supplement for the description of synoptic weather pattern near the Korean Peninsula in Sect. 2.2 in the manuscript.

Figure S2 is a supplement for the description of consistency between CMAQ- and STOPS-simulated results in Sect. 2.2 in the manuscript.

Figure S3 is a supplement for the description of PM forecasting using STOPS with the real-time AOD data from GOCI in Sect. 4.1 in the manuscript.

Figure S4 is a supplement for the description of alternative emissions used for STOPS forecasting in Sect. 4.2 in the manuscript.

Table S1. Configuration and detailed physical options for WRF simulation

Number of grids	181 × 143
Horizontal resolution	27 km
Vertical layers	33 layers
Initial data	1°× 1° NCEP Final Operational Global Analysis (FNL)
Microphysics option	WSM 3-class simple ice scheme
Radiation option	RRTM (long wave) / Dudhia (short wave) scheme
Surface layer option	Monin-Obukhov (Janic Eta) scheme
Land-surface option	Unified Noah land-surface model
PBL option	YSU scheme
Cumulus option	Kain-Fritsch (new Eta) scheme

Table S2. Same as Table S1, but for CMAQ

Meteorology	WRF
Number of grids	174 × 128
Horizontal resolution	27 km
Vertical layers	15 layers
Chemical mechanism	CB05 (gas-phase) / AERO6 (aerosol)
Chemical solver	Smvgear
Horizontal advection	Yamo
Horizontal diffusion	Multiscale
Vertical advection	WRF
Vertical diffusion	ACM2
Deposition	M3dry
Anthropogenic emissions	MIX-2010 / CAPSS 2011
Dust emission model	In-line windblown dust model

Table S3. Statistical parameters for the WRF simulation results during the entire simulation period (February 2015) at 20 observational sites. The location of each site is shown in Fig. 2 in the manuscript

Sites	Temperature			Wind Speed		
	RMSE	IOA	MBE	RMSE	IOA	MBE
S1	0.78	0.99	-0.08	1.12	0.97	0.03
S2	1.46	0.98	0.17	1.38	0.90	0.15
S3	2.49	0.90	-0.27	1.23	0.80	-0.85
S4	1.94	0.93	1.80	1.28	0.78	-0.21
S5	2.31	0.93	1.48	1.13	0.84	-0.40
S6	2.31	0.93	1.04	1.89	0.91	1.49
S7	2.48	0.96	-1.46	1.96	0.77	1.43
S8	2.58	0.93	-1.58	1.61	0.87	1.25
S9	1.40	0.94	1.39	1.19	0.86	1.12
S10	1.42	0.95	1.41	1.87	0.91	1.21
S11	2.02	0.97	-1.06	2.03	0.75	1.45
S12	2.70	0.78	-2.35	1.34	0.92	0.94
S13	2.11	0.94	1.24	1.24	0.88	0.85
S14	1.59	0.95	1.01	2.07	0.93	1.46
S15	2.67	0.89	-2.29	2.37	0.76	1.90
S16	1.39	0.98	0.43	1.59	0.89	0.90
S17	2.48	0.84	-1.71	1.98	0.74	1.36
S18	1.60	0.96	-1.09	2.64	0.72	1.27
S19	1.58	0.95	1.17	2.03	0.82	1.02
S20	1.12	0.96	0.98	1.59	0.89	0.90
Average	1.92	0.93	0.01	1.68	0.85	0.86

Table S4. The averaged friction velocity (u_*) in three land cover categories and threshold friction velocity values ($u_{*_{ti,j}}$) for each land cover category used in CMAQ_Dust simulation

Land Cover Categories	u_*	$u_{*_{ti,j}}$ (CMAQ_Dust)
Shrubland	0.23	1.54
Mixed Shrubland-Grassland	0.16	0.55
Barren or Sparsely vegetated	0.18	0.65

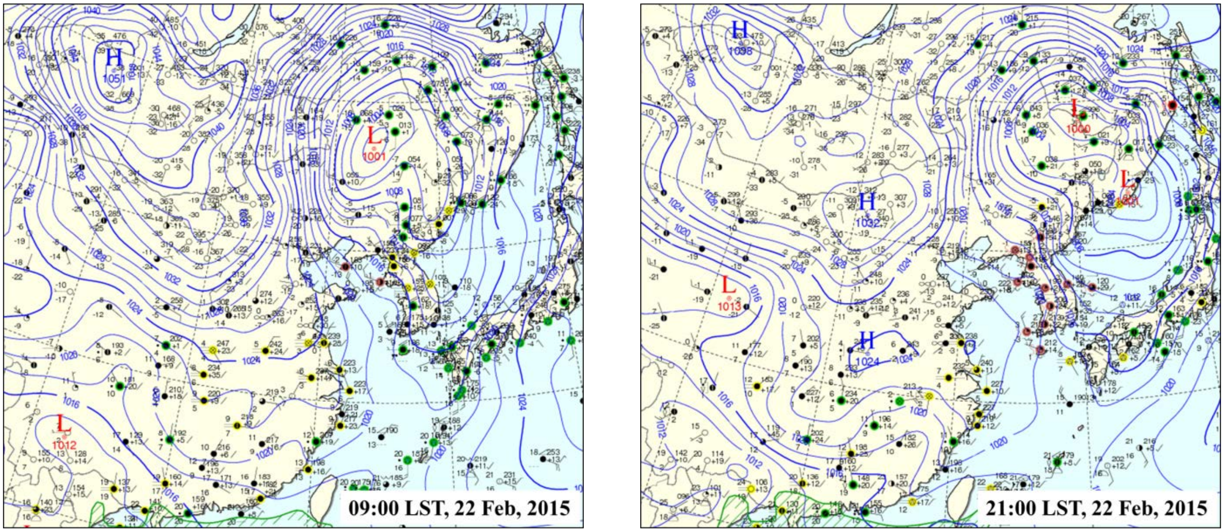

Figure S1. Surface-level synoptic weather chart near the Korean Peninsula on 22 February in 2015, which is the first day of the Asian dust event in this study.

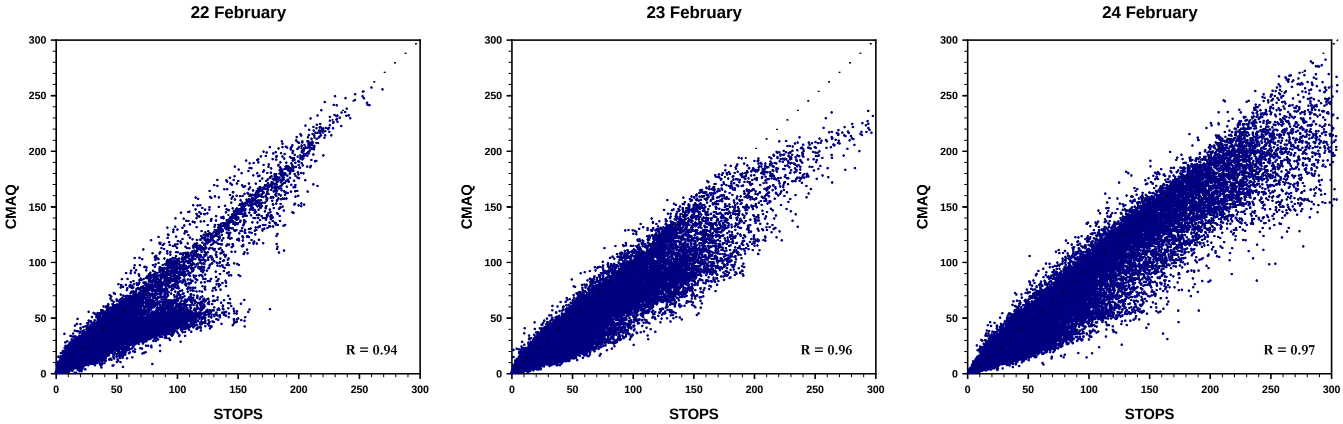

Figure S2. Scatter plots between STOPS- and CMAQ-simulated PM_{10} concentrations during the Asian dust events (22-24 February, 2015). The correlation coefficients (R) appear in the bottom-right of each plot.

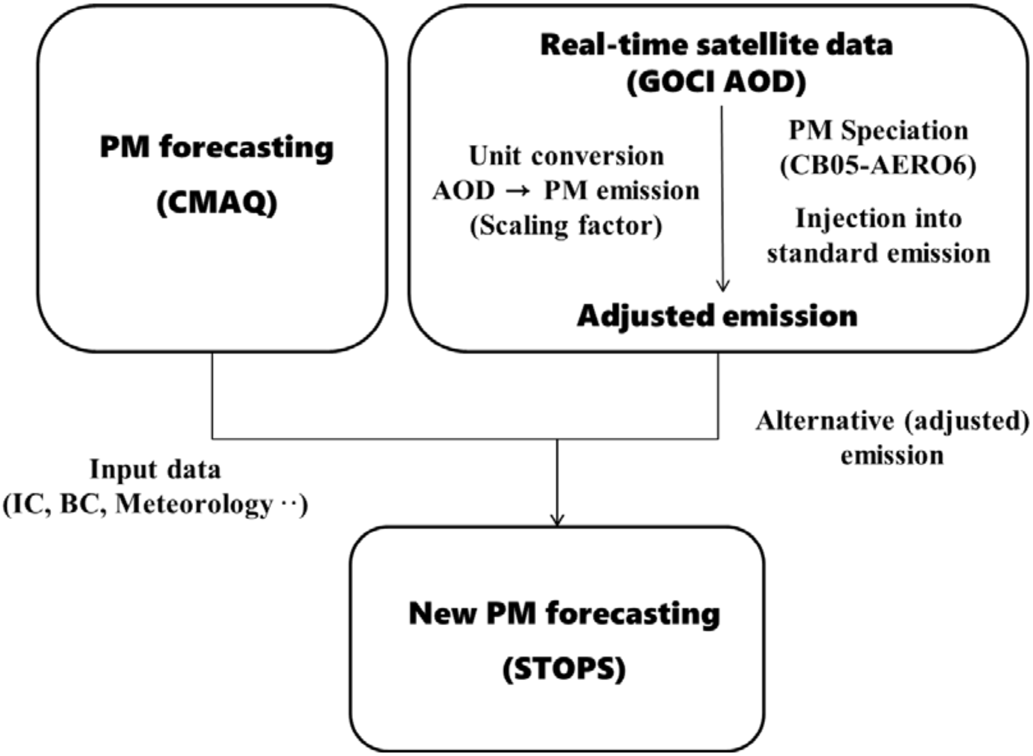

Figure S3. Schematic flowchart describing the procedure of the new PM forecasting using STOPS with the real-time AOD data from GOCI.

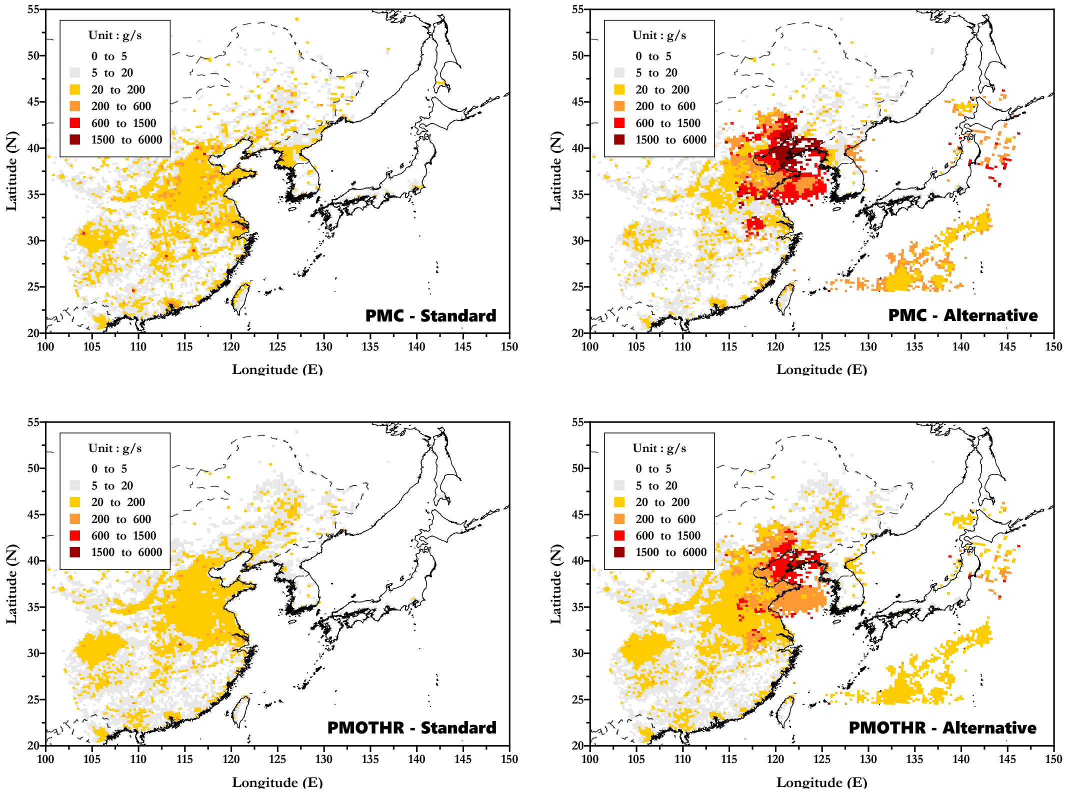

Figure S4. Difference between the emission rates (grams second^{-1}) of standard and alternative emissions (to represent enhanced GOCI AOD) data. The PMC and PMOTHR denote coarse and unspeciated fine particles, respectively.