ABSTRACT In this study, we investigate the meteorological characteristics and the effect of local emissions during high PM$_{10}$ concentrations in the Seoul Metropolitan Area (SMA) by utilizing data from a high-resolution urban meteorological observation system network (UMS-Seoul) and The Air Pollution Model (TAPM). For a detailed analysis, days with PM$_{10}$ concentrations higher than 80 μg m$^{-3}$ for daily average PM$_{10}$ concentration (classified as unhealthy by the Korean Ministry of Environment) in the Seoul Metropolitan Area (SMA) were classified into 3 Cases. Case I was defined as when the prevailing effect was from outside the SMA. Case II was defined as when the prevailing effect was a local effect with outside. Case III was defined as when the prevailing effect was local. Overall, high PM$_{10}$ concentrations in the SMA mostly occurred under weak migratory anticyclone systems over the Korean Peninsula during warm temperatures. Prior to the PM$_{10}$ concentration reaching the peak concentration, the pattern in each case was distinctive. After peak concentrations, however, the pattern for the 3 cases became less distinct. This study showed that nearly 50% of the high PM$_{10}$ concentrations in the SMA occurred in spring and were governed by the conditions for Case II more than these for Cases I and III. In spring, the main sources of the high PM$_{10}$ concentrations in the SMA were local emissions due to the predominance of weak winds and local circulation. The simulation showed that the non-SMA emissions were about 63 to 73% contribution to the spring high PM$_{10}$ concentrations in the SMA. Specifically, local point sources including industrial combustion, electric utility, incineration and cement production facilities scattered around the SMA and could account for PM$_{10}$ concentrations more than 10 μg m$^{-3}$ in the SMA.

KEY WORDS High PM$_{10}$ concentration, Korea, Local emission sources, Local meteorological circulation, Long-range transboundary processes

1. INTRODUCTION

The Seoul metropolitan area (SMA) is a mega city with an area of approximately 11,827 km$^2$ and a population of 26.1 million inhabitants (http://kosis.kr), and is ranked as the fifth largest urban area population in 2018 (Demographia, 2018). Recently, the number of days with serious concentrations of air pollutants exceeding the national ambient air quality standards of 100 μg m$^{-3}$ (35 μg m$^{-3}$) for 24-h
PM$_{10}$ (PM$_{2.5}$) concentration has increased (Park et al., 2018; NIER, 2017). For example, the Korean Ministry of Environment (KMOE) issued emergency reduction actions over the SMA on 14 January 2019 when the average PM$_{2.5}$ concentration from 0000 LST (Local Standard Time; UTC + 0900) to 1600 LST was over 50 μg m$^{-3}$ and the forecast average PM$_{2.5}$ concentration for 24 h later was greater than 50 μg m$^{-3}$ (KMOE, 2017). These actions resulted in the prohibition of older diesel vehicles from entering downtown areas in the SMA and a two-shift system for permissible vehicles to enter.

Concerns about air pollution have led to the operation of a modeling system for the daily air quality prediction to support control actions within the SMA (Park et al., 2004). High PM$_{10}$ concentrations in a particular region can be caused by trapped particle emissions and/or local secondary generation of particulate matter through photochemical reactions in the atmosphere (Kim et al., 2017; Seo et al., 2017). Recent high PM$_{10}$ concentrations in Korea appear to partly result from transboundary pollution from China, but also from domestic sources originating from local emissions (Wang et al., 2018; Park et al., 2015, 2005; Jo and Kim, 2013; Kim et al., 2012). From January 1, 2016 to January 31, 2019, there were 65 occurrences of high PM$_{10}$ concentration in the SMA where the daily average PM$_{10}$ concentration without the effect of Asian dust was greater than 80 μg m$^{-3}$ (classified as unhealthy by KMOE). Higher PM$_{10}$ concentrations generally occur on warm days in spring, autumn, or winter and are believed to be closely related to warm temperatures in the atmosphere (Park, 2014). For example, in 2016, an advisory by MOE was in effect over the SMA from April 8 to 12 with a PM$_{10}$ concentration on April 9 of 241 μg m$^{-3}$.

Previous studies have investigated the occurrence of high PM$_{10}$ concentration in Korea (Park, 2016). During most episodes (Seo et al., 2018), the Korean peninsula is influenced by anticyclonic circulation with warm and humid air with stagnant conditions, based on surface and upper atmosphere maps during high PM$_{10}$ concentration episodes (Seo et al., 2017; Park, 2016). Under these conditions, the stable atmosphere with an inversion layer and thick fog is unfavorable for transport and dispersion of particulate matter (Seo et al., 2018). Other mechanisms for high PM$_{10}$ concentrations, such as transboundary processes from local sources, also need to be considered for a comprehensive understanding of the occurrence of long-lasting high PM$_{10}$ concentration episodes (Park et al., 2018; Park, 2016). Understanding the processes and impacts of both transboundary processes and local emissions is still an important issue for developing effective emission abatement strategies in Korea.

The main objective of this study was to analyze the meteorological characteristics and PM$_{10}$, PM$_{2.5}$, NO$_2$, and SO$_2$ concentrations to assess the effect of local emissions during high PM$_{10}$ concentration. In this study, high PM$_{10}$ concentration in the SMA is defined as when the daily average PM$_{10}$ concentration (without the effect of Asian dust) is greater than 80 μg m$^{-3}$. All results representing the SMA refer to central Seoul, the capital city of South Korea.

2. MATERIALS

2.1 Meteorological and Air Quality Data

Hourly mean air quality data such as PM$_{10}$, PM$_{2.5}$, SO$_2$, NO$_2$, and O$_3$ concentrations at Seoul and Baengnyeong-do (BND) stations were used (http://www.airkorea.or.kr) (Fig. 1). Seoul station is located near the City Hall and at the center of Seoul, and is surrounded by many line and area sources (cross mark in Seoul in Fig. 1). The BND station, one of national background air quality monitoring station, has few nearby anthropogenic emission sources (cross mark in BN in Fig. 1). It is located about 208 km west of Seoul Station (Fig. 1). Surface meteorology and boundary structure were obtained from the surface energy balance observation tower and the lidar-type ceilometer installed at Jungnang Station (diamond in Seoul in Fig. 1; Park, 2018; Park et al., 2017). The station is located 9 km east of Seoul Station. Temperature, relative humidity, wind speed and direction, downward solar radiation, and 3-dimensional sonic anemometer sensors are installed on a surface energy balance tower. Friction velocity is computed by applying an eddy covariance method using 30-min block averaging with 10-Hz meridional, zonal, and vertical wind speed (Kwon et al., 2014; Park et al., 2014). A ceilometer (model: CL51, manufacturer: Vaisala) gives vertical profiles (10-m vertical resolution) of two-way attenuated backscatter due to atmospheric aerosols by means of 910 nm wavelength laser. Backscatter exceeding $400 \times 10^{-8}$ sr$^{-1}$ m$^{-1}$ was excluded to remove the effects of high backscatter media such as clouds (Park, 2018).

2.2 High PM$_{10}$ Concentrations Classification

When daily mean PM$_{10}$ concentration in Seoul was
higher than 80 μg m\(^{-3}\), the day was classified as a high PM\(_{10}\) concentration. As shown in Table I, there were 65 high PM\(_{10}\) concentration days in the SMA during the period from January 1, 2016 to January 31, 2019. In order to compare the characteristics of high PM concentration, the high PM concentrations days were categorized into 3 Cases: Case I was defined as when the PM\(_{10}\) concentration in BND area was higher than that in the SMA, and the PM\(_{10}\) (PM\(_{2.5}\)) concentration at BND Station was higher than 100 μg m\(^{-3}\) (50 μg m\(^{-3}\)). The high PM\(_{10}\) in this case could be considered to be transported from mainly long-range transboundary processes; Case II was defined as when the PM\(_{10}\) concentration in BND was lower than that in the SMA, and the PM\(_{10}\) (PM\(_{2.5}\)) concentration was higher than an annual mean of 38 μg m\(^{-3}\) (22 μg m\(^{-3}\)) at BND. As mentioned, because the BND station is far from direct emission sources, the mean PM concentration in BND was much lower than that in Seoul. Thus, annual mean concentrations at BND were considered to discriminate local-emission from contributions from long-range transboundary processes. High PM concentrations in this case could be result from both local and long-range transboundary processes; Case III was defined as when the PM\(_{10}\) (PM\(_{2.5}\)) concentration at BND was lower than one-half of that in the SMA. High PM concentrations in this case could be considered to be emitted from mainly local sources. Annual mean PM concentrations at BND rather than the national standards would be enough to reveal

| Season | Case I | Case II | Case III | Total |
|--------|--------|---------|----------|-------|
| Spring | 4 (8)  | 26 (19) | 2 (5)    | 32 (32)|
| Autumn | 2 (2)  | 2 (–)   | 4 (2)    |       |
| Winter | 8 (13) | 17 (13) | 4 (3)    | 29 (29)|

Monitoring data for PM\(_{2.5}\) was missing for 2 days.
contrasts between local-emission and long-range transboundary processes in the SMA.

According to the above criteria, around 70% (45) of all high PM10 concentration days was classified as Case II, while 18% (12) and 12% (8) were Case III and I, respectively (Table 1). Also, 49% and 45% were occurred in spring and winter, respectively, while only 6% was occurred in autumn. For investigation of detailed features, 3 events for all cases were selected: one spring and two winter (no event in autumn) were selected for Case I. One event in each season (spring, autumn, and winter) for Case II and III was selected, respectively. Table 2 shows the daily mean PM10, PM2.5, NO2, SO2, CO, and O3 concentrations at BND and Seoul Stations for all high PM10 concentration days.

### 3. CHARACTERISTICS OF AIR POLLUTANTS

Time series of PM10, PM2.5, NO2, and SO2 concentration during the 7 days centered on the high concentration day at SMA and BND are shown in Fig. 2. Median concentrations and ratios of the 7-day mean concentration to the monthly mean concentration for each case and each pollutant are summarized in Tables 3 and 4, respectively. The detailed features of the temporal variation were analyzed for each case.

#### Case I: The PM10 and PM2.5 concentrations in Seoul showed a lag time 6 to 12 h for the maximum concentration compared to those in BND during high PM10 concentration for all cases. The maximum PM10 concentration of 111 μg m\(^{-3}\) (180 μg m\(^{-3}\)) in Seoul was much lower than 386 μg m\(^{-3}\) (299 μg m\(^{-3}\)) at BND for the spring (winter_a) case (Fig. 2). Because there were few direct NO2 sources at BND, NO2 concentrations at BND were much lower than in the SMA during the study period. The SMA had nearly the same SO2 or lower concentration than BND. The median values in the SMA were nearly the same as in BND for PM10, PM2.5, and SO2 concentrations, while much higher than BND for NO2 concentration (Fig. 3; Table 3). The ratio of the 7-day mean concentration to monthly mean concentration showed high values for PM10 and PM2.5 concentration, while nearly the same values for NO2 and SO2 at both the SMA and BND (Table 4).

#### Case II: The PM10 and PM2.5 concentrations in the SMA were nearly the same as concentrations in BND. NO2 concentration in BND was much less than that in the SMA, while SO2 concentration in the SMA was higher than that in BND, especially in winter (Fig. 2). In autumn, high PM10 concentrations in BND were observed later than in the SMA by about 6 h, and high NO2 concentration in BND were recorded on 18–19 November 2016. These implied that air parcels in BND might be
Figure 2. Time series of PM\(_{10}\) (first panel), PM\(_{2.5}\) (second panel), NO\(_2\) (third panel), and SO\(_2\) (fourth panel) concentrations in the SMA and BND during the 7 consecutive days around the maximum concentration for each case. High PM\(_{10}\) concentration days are shown by green shading.
advected from the SMA. The median values in the SMA were similar to or higher than BND for PM$_{10}$, PM$_{2.5}$, and SO$_2$ concentrations, while much higher than BND for NO$_2$ concentration (Table 3). The ratios of the 7-day mean concentration to the monthly mean concentration were more or less unity except for high for PM concentrations in both areas in winter and low SO$_2$ concentrations at BND in autumn and winter (Table 4).

Case III: The PM$_{10}$ and PM$_{2.5}$ concentrations in the SMA were nearly the same as concentrations at BND prior to the high PM concentration period in the SMA. However, PM concentrations in the SMA increased, but those in BND sharply decreased (Fig. 2). These implied that prior to the peak of PM concentration in the SMA, the SMA might be influenced by air masses that passed the BND region. But after the peak, SMA and BND might be influenced by different air masses. So high PM concentration in the SMA was mainly contributed by local emission. The NO$_2$ and SO$_2$ concentrations in BND were much less than concentrations in the SMA, except for SO$_2$ concentrations in winter. The median concentrations in the SMA were much higher than those
Table 3. Median PM$_{10}$, PM$_{2.5}$, NO$_2$, and SO$_2$ concentrations in the SMA and BND for the 3 cases.

| Case | Season | Season (Date) | PM$_{10}$ (μg m$^{-3}$) | PM$_{2.5}$ (μg m$^{-3}$) | NO$_2$ (ppm) | SO$_2$ (ppm) |
|------|--------|---------------|-------------------------|-------------------------|-------------|-------------|
| I    | Spring | Apr 27–May 3, 2017 | 67.0$^{(1)}$ | 29.5$^{(1)}$ | 0.034$^{(1)}$ | 0.005$^{(1)}$ |
|      | Winter_a | Jan 17–23, 2018 | 51.0 | 32.0 | 0.043 | 0.004 |
|      | Winter_b | Jan 10–16, 2019 | 84.5 | 58.0 | 0.053 | 0.005 |
| II   | Spring | Mar 9–15, 2018 | 35.0$^{(1)}$ | 23.0$^{(1)}$ | 0.033$^{(1)}$ | 0.004$^{(1)}$ |
|      | Autumn | Nov 15–21, 2016 | 54.0 | 32.0 | 0.050 | 0.003 |
|      | Winter | Dec 19–25, 2018 | 48.0 | 29.0 | 0.052 | 0.005 |
| III  | Spring | Mar 23–29, 2018 | 78.0$^{(1)}$ | 51.0$^{(1)}$ | 0.040$^{(1)}$ | 0.003$^{(1)}$ |
|      | Autumn | Nov 3–9, 2018 | 44.5 | 30.0 | 0.046 | 0.004 |
|      | Winter | Feb 1–7, 2017 | 43.0 | 28.0 | 0.041 | 0.004 |

1)$^{(1)}$SMA, 2)$^{(2)}$BND

Table 4. Ratio of the 7-day average concentration (calculated as three days before and three days after the maximum concentration) to the monthly average concentration for the 3 cases.

|        | Spring   | Autumn   | Winter   |
|--------|----------|----------|----------|
|        | Case I   | Case II  | Case III | Case I   | Case II  | Case III | Case I (a) | Case I (b) | Case II  | Case III |
| PM$_{10}$ | 1.24$^{(1)}$ | 0.94 | 1.76 | 1.17 | 1.02 | 1.33 | 1.53 | 1.21 | 1.18 |
| PM$_{2.5}$ | 1.22$^{(1)}$ | 0.98 | 1.84 | 1.15 | 1.23 | 1.36 | 1.78 | 1.29 | 1.29 |
| NO$_2$ | 0.95$^{(1)}$ | 1.06 | 1.26 | 1.16 | 1.09 | 1.15 | 1.19 | 1.35 | 1.09 |
| SO$_2$ | 1.30$^{(1)}$ | 1.02 | 1.31 | 1.03 | 0.99 | 1.06 | 1.10 | 1.29 | 1.00 |

1)$^{(1)}$SMA, 2)$^{(2)}$BND

in BND for PM$_{10}$, PM$_{2.5}$, and NO$_2$ (Fig. 3; Table 3). The ratios of the 7-day mean concentration to the monthly mean concentration were larger than unity in spring, but more or less unity in autumn, and winter (Table 4).

4. METEOROLOGICAL CHARACTERISTICS

4.1 Synoptic Analyses

To identify the impacts of meteorological conditions on PM concentrations, surface weather charts near the Korean Peninsula at 0900 LST for selected event days shown in Fig. 4 were investigated.

Case I: A relatively small-scale high pressure system affected the entire Korean Peninsula for all event days. It started from northwest of the Korean Peninsula, and was separated from the periphery of the Siberian High, especially in winter (Fig. 4b, c). A small anticyclonic system, originated from rear-side of the upper trough, passed through the Korean peninsula from northwest to southeast in spring (Fig. 4a).

Case II: A weak anticyclone system, separated from the high pressure system centered on the Japanese Islands, was stagnant over the Korean Peninsula in spring and autumn (Fig. 4d, e). The SMA was affected by a small-scale high pressure system originating from the Siberian High in winter (Fig. 4f). Due to the position of
the weak high pressure system near the Korean Peninsula in autumn and winter, weak easterly winds became dominant in the SMA. The easterly wind might be evidence for advection from SMA to BND in autumn and winter, and the high NO2 concentration in BND (Fig. 2).

Case III: The synoptic meteorological patterns in spring and winter were similar to that for the Case II autumn case (Fig. 4g, h). A small-scale high pressure system, separated from the southern part of the Korean Peninsula, was stationary over the SMA in winter (Fig. 4i).

Overall, most high PM concentration episodes in the SMA were accompanied by weak migratory anticyclone systems over the Korean Peninsula. The horizontal extent of surface pressure near the Korean Peninsula for all cases were very small, implying that the synoptic system near the SMA was weak and stagnant during the event periods, as previous studies have shown (Park, 2018; Seo et al., 2017).

### 4.2 Surface Meteorology and Boundary-Layer Structure

In order to understand the effect of surface meteorology and boundary-layer structure on PM10 and PM2.5 concentrations near the surface, time series of meteorological variables (Fig. 5) and the time-height cross section of attenuated backscatter (Fig. 6) measured at Jungnang Station were analyzed (Fig. 1).

Case I: Strong south-westerlies were dominant for the spring case, but weak winds with local circulation were dominant for the winter case (Fig. 5). Air temperature showed an increasing trend for the spring case, and had nearly the same trend but was much warmer than the climatological mean (–2.8 to –2.2°C) for the winter case (http://www.kma.go.kr). Solar radiation during the high PM10 episode was lower than on the antecedent clear days. It was due to high scatter of incident solar radiation by atmospheric aerosol (Barmpadimos et al., 2011).
Fig. 5. Time series of observed (red line and open circle) and simulated (blue dashed line and closed rectangle) wind speed (first row), wind direction (second row), net solar radiation (third row), friction velocity (fourth row), air temperature (fifth row), and relative humidity (sixth row) at Jungnang station during the 7 consecutive days around the maximum concentration for each case. High PM$_{10}$ concentration days are shown by green shading.
Fig. 6. Time-height cross section of attenuated backscatter and time series of mixing layer height at Jungnang station during the 7 consecutive days around the maximum concentration for each case (a-i).
Meteorology and Local Emission during High PM Concentration in SMA

precipitation event of 3.3 mm was recorded on January 22, 2018. The friction velocity indicated mechanical turbulence and was proportional to wind speed. Weak friction velocity meant the poor vertical and horizontal diffusion (Li et al., 2018). Attenuated backscatter near the surface tended to be proportional to the PM10 concentration (Fig. 2). The aerosol layer formed at 300–700 m in the morning, and descended to the surface in the evening on April 30, 2017 in spring. For the winter_a case, an aerosol layer formed near about 1 km height at 0000 LST on January 20, 2018. The aerosol layer combined with the evolving mixing layer in the morning, and exhibited the maximum PM10 concentration near surface in the early evening. For the winter_b case, the aerosol layer formed near the surface and in the overlying residual layer, respectively, at 0000 LST on January 11, 2019. The upper aerosol intruded into the mixing layer two times: from 1 km height in the morning on January 12, and from 500 m height at 0300 LST on January 13, 2019. As a result, the maximum PM10 concentration at the surface occurred during the late evening on January 14, 2019.

Case II: Relatively strong westerlies were dominant for the spring case like Case I, but very weak north-easterly winds accompanied by local circulation were dominant for the autumn and winter cases (Fig. 5). The air temperature was much warmer than the climatology (3.9 to 5.9°C for the spring case; 5.6 to 7.0°C for the autumn case; −0.18 to 0.4°C for the winter case). Solar radiation on the high PM10 concentration day was lower than on antecedent clear days. Weak northeasterly wind occurred in the morning and the aerosol layer at 1 km height intruded into the mixing layer in the afternoon on March 12, 2018 for the spring case (Park and Chae, 2018). For the autumn case, the aerosol layer at 1.5 km height on November 16, 2016 intruded into the mixing layer in the next afternoon (Fig. 6). After a weak rain in the late evening, PM10 concentrations decreased on the next day. For the winter case, a weak aerosol layer intruded into the surface layer on December 20, 2018 and in the morning on December 22, 2018, respectively.

Case III: Relatively strong westerlies were dominant for the spring and winter cases, but weak northeasterly winds were dominant for the autumn case. Differing PM10 concentrations in the SMA and BND was accompanied by southwesterly winds for the spring case, and northeasterly winds for the autumn and winter cases (Fig. 5). Air temperature showed an increasing trend for the spring case, and was warmer than the climatology (9.3 to 10.4°C for the autumn case, and −2.4 to −0.9°C for the winter case). The aerosol layer was trapped in the lowest 500 m and 300 m in the morning on 25 and 26 March 2018 for the spring case, respectively. There was no intrusion from the upper layer for the autumn case (Fig. 6). After the high PM10 concentration, a heavy rain event (64 mm) occurred on the next day. The aerosol layer was trapped in the lower 300 m under weak wind and a weak aerosol layer intruded from 1 km height on February 4, 2017 for the winter case.

5. TAPM MODELING

TAPM employed in this study is composed of two prognostic subsections: meteorological and chemical modules (Hurley, 2008). The TAPM meteorological module predicts gridded three-dimensional meteorology and air pollutant concentrations. The meteorological component of TAPM is an incompressible, non-hydrostatic, primitive equation model with a terrain-following vertical coordinate for three-dimensional simulations. The model solves the primitive meteorological equations: momentum, temperature, water vapor specific humidity, and cloud/rain/snow components. It includes parameterizations for cloud/rain/snow micro-physical processes, turbulence closure, urban/vegetation canopy and soil, and radiative fluxes (Hurley, 2008). The chemical component of TAPM consists of an Eulerian grid-based set of prognostic equations for both gas and particulate components. TAPM includes gas phase photochemical reactions based on semi-empirical mechanisms called the generic reaction set (GRS) of Azzi et al. (1992) with the hydrogen peroxide modification of Venkatram et al. (1997). More details on TAPM equations and descriptions can be found in Hurley (2008) and Hurley et al. (2008).

The domain for TAPM in this study was based on grids of 100 × 100 × 25 at three domains (12 km, 4 km, 2 km), centered in central Seoul (latitude 37°33.970′N and longitude 126°58.671′E). The initial conditions of the meteorological variables, LAPS (Local Administrator Password Solution) data with a resolution of 75 km × 100 km (ftp://ftp.csiro.au/TAPM) and surface and land use data (1 km × 1 km) from the USGS (United States Geological Survey) (Hurley, 2008) were used.
Nationwide average concentrations of PM$_{10}$, PM$_{2.5}$, SO$_2$, NO$_2$, CO, and O$_3$ during 3 days before the maximum PM$_{10}$ concentration were used for the initial background model conditions (Table 5). A spin-up time of 5 days was applied to minimize the initial condition influences for both surface and non-surface concentrations.

5.1 Emissions
Total and categorized (area, line, point) emissions of PM$_{10}$, NO$_x$, SO$_2$, and CO in nationwide and in the SMA for 2015 were used in this study. Total emissions were 232,794 ton y$^{-1}$, 1,000,064 ton y$^{-1}$, 351,010 ton y$^{-1}$, and 941,238 ton y$^{-1}$ nationwide and 44,385 ton y$^{-1}$, 289,431 ton y$^{-1}$, 37,223 ton y$^{-1}$, and 307,929 ton y$^{-1}$ in the SMA for PM$_{10}$, NO$_x$, SO$_2$, and CO, respectively (Table 6). PM$_{10}$ and CO emissions mostly originate from area sources, while most of the NO$_x$ emissions are from line sources and SO$_2$ emissions are from point sources (Park et al., 2004). Each of these source categories were considered and urban-scale air pollutants were accurately inventoried by taking into account of all the information on stationary sources such as stack height, stack diameter, exit velocity, and temperature. Fig. 7 illustrates the spatial distribution of total and apportioned categorized emissions for PM$_{10}$, NO$_x$, and SO$_2$ for each grid (in ton y$^{-1}$ (2 km × 2 km)$^{-1}$). Total emissions of PM$_{10}$ and NO$_x$ were found to be highly dependent on traffic densities, and the largest emission occurred in the central SMA, along expressways, and at junctions of major roads or streets. High emissions of SO$_2$ were found along the western coastal area, where higher densities of power plants and industries are located (Park et al., 2004).

5.2 Model Evaluation
In order to validate the simulated results, the simulated

PM$_{10}$ concentrations at Seoul station and simulated meteorology at Jungnang station, located 11 km from central Seoul were used. The evaluations were performed by using statistical performance measures such
as the index of agreement (IOA), root mean square error (RMSE), and correlation coefficient (R). IOA is a frequently used measure of how well the predicted variation about the observed mean is represented, with a value greater than about 0.50 considered to indicate a good prediction (Hurley et al., 2008).

Simulated meteorological variables were, by and large, well coincident with the observed ones for all cases (Fig. 5). Wind speed was slightly underestimated under strong wind, while downward solar radiation was overestimated by around 10% in spring. Nighttime temperature (relative humidity) were underestimated (overestimated) in spring. These differences were mainly due to the urban heat island (Park and Chae, 2018). TAPM exhibited some weaknesses in simulating the urban heat island over vast urban surfaces. In order to verify the performance of the model, the mean bias (MB), root mean square error (RMSE), correlation coefficient (R), and index of agreement (IOA) for Case 1 Spring, Case 2 Autumn, and Case 3 Winter cases were evaluated (Table 7). The results showed that the meteorology (temperature, wind speed, and wind direction) were reasonably well simulated for all selected cases. For PM$_{10}$ concentration, the IOAs were above 0.6, indicating the simulated PM$_{10}$ concentration was in good agreement with observed concentration (Hurley et al., 2008). The IOAs in this

![Spatial distribution of air pollutant emissions in the SMA. Total emissions for PM$_{10}$ (a), NO$_x$ (b) and SO$_2$ (c); area source emissions for PM$_{10}$ (d), NO$_x$ (e) and SO$_2$ (f); line source emissions for PM$_{10}$ (g), NO$_x$ (h) and SO$_2$ (i); point source emissions for PM$_{10}$ (j), NO$_x$ (k) and SO$_2$ (l). Black-dotted line represents the boundary of SMA.](image)
study were found to be generally better than those in recent similar studies in urban areas (Park et al., 2018).

5.3 Wind and Concentration Fields

Fig. 8 shows the horizontal distribution of PM$_{10}$ with the wind vector simulated with nationwide emissions for the 3 cases. The patterns are strongly governed by the wind; upwind regions had lower concentrations while downwind regions had higher concentrations. The areas where confluence of wind occurred had higher concentrations and areas where the wind diverged had lower concentrations. The overall concentration field in spring and autumn showed an inhomogeneous horizontal distribution pattern. The highest concentration was in the western coastal area due to the high density of point sources such as power plants, industrial complexes and steel production facilities, and also high in the central SMA due to the high density of line sources, with somewhat higher PM$_{10}$ concentrations in the southeast due to the presence of cement manufacturing facilities. The PM$_{10}$ concentration in winter was higher in the southern area due to the prevailing northwesterly wind and showed a more homogeneous horizontal distribution pattern due to stronger winds in winter. The concentration plu-
me for Case I was also broader than for Case II and Case III due to relatively strong northwesterly wind (Park et al., 2004).

Fig. 9 shows the horizontal distribution of PM$_{10}$ with the wind vector simulated without SMA emissions for the 3 cases. The overall concentration field showed a homogeneous horizontal distribution pattern except for the highest PM$_{10}$ concentrations in the western coastal area. It should be noted that point sources including industrial combustion, electric utility, incineration, cement manufacturing, and residential heating facilities in the satellite cities located to the northeast of the SMA were important local sources that can account for at least 10 μg m$^{-3}$ in the SMA for Case II in spring and Case III in autumn. The temporal variation of simulated PM$_{10}$ with nationwide emissions showed higher diurnal variation and higher concentrations compared to simulated concentration without SMA emissions. PM$_{2.5}$ concentration showed less diurnal variation and lower concentrations compared to PM$_{10}$ concentration.

5.4 Assessment of Local Emissions during High PM$_{10}$ Concentration

The simulated average surface concentrations were used to assess the fractional contribution of total local emissions of PM$_{10}$, PM$_{2.5}$, NO$_x$, and SO$_x$ in the SMA during high PM concentration for each case. The contribution of local emission was assessed by analyzing the
Table 8. Fractional contribution of local emissions on the average concentrations in the SMA for the 3 cases (value considering two power plants shown in parenthesis).

| Case | Season | Nationwide | SMA | Reduced Conc. | Fraction (%) | PM$_{10}$ (μg m$^{-3}$) | PM$_{2.5}$ (μg m$^{-3}$) | NO$_2$ (ppb) | SO$_2$ (ppb) |
|------|--------|------------|-----|--------------|--------------|----------------|----------------|-------------|----------|
| I    | Spring | Nationwide | 76.6 | 54.9 (54.8) | 21.7 | 28.3 (28.5) | 45.3 | 29.6 (29.5) | 15.7 | 15.7 | 17.6 (14.3) | 4.9 (3.8) | 4.9 (3.8) |
|      | Winter_a | Nationwide | 76.6 | 54.9 (54.8) | 21.7 | 28.3 (28.5) | 45.3 | 29.6 (29.5) | 15.7 | 15.7 | 17.6 (14.3) | 4.9 (3.8) | 4.9 (3.8) |
|      | Winter_b | Nationwide | 59.7 | 54.9 (54.9) | 4.8 | 8.0 (8.0) | 46.1 | 42.6 (42.6) | 24.1 (24.0) | 24.1 (24.0) | 76.6 (73.6) | 27.8 (27.3) | 3.6 (3.3) |
|      | Spring | Nationwide | 47.8 | 30.3 (30.3) | 17.5 | 36.6 (36.6) | 37.1 | 24.5 (24.5) | 19.7 (18.5) | 19.7 (18.5) | 7.6 (7.6) | 7.6 (7.6) | 7.6 (7.6) |
|      | Autumn | Nationwide | 68.9 | 49.4 (49.4) | 19.5 | 28.3 (28.3) | 43.1 | 29.0 (29.0) | 25.1 (24.8) | 25.1 (24.8) | 7.6 (7.6) | 7.6 (7.6) | 7.6 (7.6) |
|      | Winter | Nationwide | 59.7 | 46.2 (46.2) | 13.5 | 22.6 (22.6) | 44.4 | 34.4 (34.4) | 28.3 (27.8) | 28.3 (27.8) | 6.3 (6.3) | 6.3 (6.3) | 6.3 (6.3) |
|      | Spring | Nationwide | 66.2 | 48.3 (48.2) | 17.9 | 27.0 (27.2) | 56.4 | 43.0 (42.9) | 20.9 (20.1) | 20.9 (20.1) | 8.4 (8.4) | 8.4 (8.4) | 8.4 (8.4) |
|      | Autumn | Nationwide | 40.0 | 29.6 (29.6) | 10.4 | 26.0 (26.0) | 32.0 | 24.4 (24.4) | 18.4 (18.4) | 18.4 (18.4) | 4.3 (4.3) | 4.3 (4.3) | 4.3 (4.3) |
|      | Winter | Nationwide | 44.1 | 32.8 (32.8) | 11.3 | 25.6 (25.6) | 32.5 | 24.1 (24.0) | 22.0 (21.6) | 22.0 (21.6) | 3.7 (3.5) | 3.7 (3.5) | 3.7 (3.5) |

The difference between model results with and without SMA emission based on nationwide emissions. This approach is basically the Brute-Force Method (BFM) by zeroing-out the emission on the targeted area (Koo et al., 2009).

For Case I, the fractional contribution to the average surface concentration ranged from 8 to 28% for PM$_{10}$, 8 to 35% for PM$_{2.5}$, 28 to 74% for NO$_2$, and 33 to 52% for SO$_2$. For Case II, the fractional contribution to the average surface concentration ranged from 23 to 37% for PM$_{10}$, 23 to 34% for PM$_{2.5}$, 35 to 57% for NO$_2$, and 33 to 60% for SO$_2$. For Case III, the fractional contribution to the average surface concentration ranged from 25 to 27% for PM$_{10}$, 24 to 26% for PM$_{2.5}$, 39 to 65% for NO$_2$, and 44 to 63% for SO$_2$ (Table 8). Distinct characteristics for each case appeared only for Case I in winter with the fractional contribution to average surface concentration of local emissions of 8% for PM$_{10}$, 8% for PM$_{2.5}$, 28% for NO$_2$, and 33% for SO$_2$.

The fractional contribution of non-SMA emissions to the SMA on the average pollutant concentration for the 3 cases was analyzed. For Case I, the fraction of non-SMA emissions was 83% for PM$_{10}$, 81% for PM$_{2.5}$, 55% for NO$_2$, and 58% for SO$_2$ on average. For Case II, the fraction of non-SMA emissions was 71% for PM$_{10}$, 70% for PM$_{2.5}$, 64% for NO$_2$, and 66% for SO$_2$ on average. For Case III, the fraction of non-SMA emissions was 69% for PM$_{10}$, 68% for PM$_{2.5}$, 59% for NO$_2$, and 61% for SO$_2$ on average.
for PM$_{2.5}$, 54% for NO$_{2}$, and 43% for SO$_{2}$ on average. For Case III, the fraction of non-SMA emissions was 74% for PM$_{10}$, 75% for PM$_{2.5}$, 48% for NO$_{2}$, and 44% for SO$_{2}$ on average (Table 9).

The fractional contribution for the average concentration in the SMA during high PM$_{10}$ concentrations was consistent for the 3 cases except winter in Case I. It should be noted that the classification into the 3 cases in this study was based on the daily average PM$_{10}$ and PM$_{2.5}$ concentrations, not meteorological characteristics. Before the maximum PM$_{10}$ concentration at Seoul, the temporal variation for each case showed special features (Fig. 2). The PM concentrations in BND exhibited their peaks prior to those in SMA for Case I, while the latter exhibited their peaks prior to the former for Case III. Whereas the PM concentrations in both BND and Seoul exhibited their peaks at similar time for Case II.

Especially for Case II, polluted air masses from the outside of SMA were transported into the SMA by westerly (in spring) and northerly (in autumn and winter) winds before the high PM$_{10}$ concentration events (Fig. 5). During the high PM concentration event periods in spring and autumn, stagnant synoptic systems over the Korea Peninsula, due to the surrounding pressure patterns (e.g., the high pressure system over Japan), hindered the horizontal transport and vertical dispersion of pollutants. Thus, air pollutants were accumulated, and their concentrations increased in the SMA. During the events in winter, a small-scale high pressure system was located over the Korean Peninsula. Easterly winds were too weak to disperse air pollutants effectively (Figs. 4-6).

Although the TAPM experiments indicated larger impacts of non-SMA emission and long-range transport processes on the high PM concentrations in the SMA (e.g., 71% for PM$_{10}$, 70% for PM$_{2.5}$, and 54% for NO$_{2}$) (Table 9), this does not mean that the contribution of local emissions was negligible, because local emissions still contributed between 30% and 50% to the high concentrations of air pollutants. This suggests that both long-range transport processes and locally emitted pollutants play a role as a contributor to high PM$_{10}$ concentration events in the SMA.

The Dangjin and Taean power plants located on the coast southwest of the SMA have been identified as large point sources in South Korea (Park et al., 2018). To assess the contribution of these two power plants to the SMA, the results for simulated average surface concentrations from emissions without emissions from the SMA and the two power plants in nationwide emissions were used. For all Cases, the fractional contributions to the SMA were negligible for PM$_{10}$ and PM$_{2.5}$, and about 1% for NO$_{2}$, and 6%, 5%, and 4% for Case I, Case II, and Case III, respectively. The contribution of power plant emissions to the SMA for PM$_{10}$ and PM$_{2.5}$ in this study can be neglected. This shows that the SMA is located at an upwind location from the large two power plants during the high PM$_{10}$ concentrations. But PM$_{10}$ and PM$_{2.5}$ concentrations at downwind locations about 30 km distance from the power plant were reduced by 6% and 4%, respectively, similar to the estimate in previous studies (Park et al., 2018). This implies that local emissions from line sources in the SMA are a major source of PM$_{10}$.

### 6. CONCLUSIONS

The results of this study have shown that Case I in winter was influenced by long range transboundary processes. The PM$_{10}$ and PM$_{2.5}$ concentrations in the SMA showed a lag time 6 to 12 h for the maximum concentration compared to BND. Case III was influenced by the accumulation of local and neighboring (non-SMA) emissions by atmospheric stagnation. After the PM$_{10}$ and PM$_{2.5}$ concentrations in the SMA increased, concentrations in BND sharply decreased. Case II was a mixed case between Case I and Case III with local circulation. The PM$_{10}$ and PM$_{2.5}$ concentrations in the SMA were nearly the same as concentrations in BND. The maxi-

---

**Table 9. Fractional contribution of non-SMA emissions on average concentrations in the SMA for the 3 cases (value considering two power plants shown in parenthesis).** (unit: %)

| Case  | Season | PM$_{10}$ | PM$_{2.5}$ | NO$_{2}$ | SO$_{2}$ |
|-------|--------|-----------|-----------|---------|---------|
| Case I| Spring | 72 (72)   | 65 (65)   | 26 (24)  | 49 (38)  |
|       | Winter a | 84 (84)   | 85 (85)   | 68 (67)  | 58 (53)  |
|       | Winter b | 92 (92)   | 92 (92)   | 72 (72)  | 68 (65)  |
|       | Avg.    | 83 (83)   | 81 (81)   | 55 (54)  | 58 (52)  |
| Case II| Spring | 63 (63)   | 66 (66)   | 43 (41)  | 41 (31)  |
|        | Autumn | 72 (72)   | 67 (67)   | 54 (54)  | 43 (41)  |
|        | Winter | 77 (77)   | 78 (78)   | 66 (64)  | 46 (43)  |
|        | Avg.   | 71 (71)   | 70 (70)   | 54 (53)  | 43 (38)  |
| Case III| Spring | 73 (73)   | 76 (76)   | 36 (34)  | 38 (30)  |
|        | Autumn | 74 (74)   | 76 (76)   | 47 (47)  | 37 (37)  |
|        | Winter | 74 (74)   | 74 (74)   | 61 (60)  | 56 (53)  |
|        | Avg.   | 74 (74)   | 75 (75)   | 48 (47)  | 44 (40)  |
imum PM concentrations in BND occurred 6 h later than in the SMA by air parcels advected from the SMA.

Our study shows that the high PM$_{10}$ concentrations in the SMA were mostly governed by the conditions for Case II (45 among total 65 occurrence days) compared to for Case I (12 days) and Case III (8 days) and that nearly 50% occurred in spring (total 32 for Case I, 26 for Case II and 2 for Case III). Prior to the PM$_{10}$ concentration reaching the peak concentration, the pattern in each case was distinct. After that, the pattern for the 3 cases became less distinct, and the meteorological conditions were characterized by local circulation systems with weak wind, particularly in spring. The classification into the 3 cases in this study was based on the daily average PM$_{10}$ and PM$_{2.5}$ concentrations, not meteorological characteristics. Therefore, the fractional contribution to the average concentration in the SMA during the high PM$_{10}$ concentration, except for winter, for Case I dominated by the Siberian high pressure system, was consistent among the 3 cases. This implies that the local and neighboring (non-SMA) emissions during high PM$_{10}$ concentration in the SMA particularly in spring were the major sources rather than emissions from long-range transboundary processes and most of the non-SMA contribution to PM$_{10}$ and PM$_{2.5}$ resulted from neighboring emissions. Particularly, approximately 1600 stacks from combustion sources in the SMA and mostly industrial sources including electric industrial combustion, electric utility, incineration, cement manufacturing, and residential heating in the satellite cities located to the southeast and east of the SMA were important sources of local emissions in addition to mobile sources. The fractional contributions to high PM$_{10}$ and PM$_{2.5}$ in the SMA at upwind location from two large power plants were negligible compared to about 6% and 4% contributions to PM$_{10}$ and PM$_{2.5}$ in the other areas at downwind locations.

High PM$_{10}$ concentrations in the SMA in winter mainly resulted from long-range transboundary processes by the Siberian high pressure system. It will be valuable to confirm the fractional contribution to the SMA from long-range transboundary processes in future studies using other research approaches.

**ACKNOWLEDGEMENT**

This work was funded by the Korea Meteorological Administration Research and Development Program under Grant KMI2018-05310. All meteorological data used in this study were jointly provided by the Korea Meteorological Administration (Weather Information Service Engine Project), Hankuk University of Foreign Studies, and National Institute of Meteorological Sciences. We also thank National Institute of Environmental Research for providing Korean emission data from the Clean Air Policy Support System (CAPSS). We would like to acknowledge Jung-Hun Woo at Konkuk University for providing the aggregated Korean emission data by major sectors.

**REFERENCES**

Azzi, M., Johnson, G.M., Cope, M. (1992) An introduction to the generic reaction set photochemical smog mechanism. Proceedings of the 11th International Clean Air and Environment Conference, Brisbane, Clean Air Society of Australia & New Zealand.

Barmadimos, I., Hueglin, C., Keller, J., Henne, S., Prevot, A.S.H. (2011) Influence of meteorology on PM$_{10}$ trends and variability in Switzerland from 1991 to 2008. Atmospheric Chemistry and Physics 11, 1813–1835, DOI: 10.5194/acp-11-1813-2011.

Demographia (2018) Demographia world urban areas, built up urban areas or world agglomerations, 14th edit, Demographia, Illinois, USA, 118pp. http://demographia.com/db-worldua.pdf.

Hurley, P.J. (2008) TAPM V4. Part 1: Technical Description, CSIRO Marine and Atmospheric Research Paper No. 25, 59pp.

Hurley, P.J., Mary, E., Ashok, L. (2008) TAPM V4. Part 2: Summary of Some Verification Studies. CSIRO Marine and Atmospheric Research Paper No. 26, 31pp.

Jo, H.-Y., Kim, C.-H. (2013) Identification of long-range transported haze phenomena and their meteorological features over Northeast Asia. Journal of Applied Meteorology and Climatology 52, 1318–1328, DOI: 10.1175/JAMC-D-11-0235.1.

Kim, C.-H., Park, S.-Y., Kim, Y.-J., Chang, L.-S., Song, S.-K., Moon, Y.-S., Song, C.-K. (2012) A numerical study on indicators of long-range transport potential for anthropogenic particulate matters over northeast Asia. Atmospheric Environment 58, 35–44, DOI: 10.1016/j.atmosenv.2011.11.002.

Kim, H.C., Kim, E., Bae, C., Cho, J.H., Kim, B.U., Kim, S. (2017) Regional contributions to particulate matter contribution in the Seoul Metropolitan Area, Korea: Seasonal variation and sensitivity to meteorology and emission inventory. Atmospheric Chemistry and Physics 17, 10315–10332, DOI: 10.5194/acp-17-10315-2017.

KMOE (The Ministry of Environment in Korea) (2017) Press report (July 26): Reduction effect of shut down of old power plants during June 2017, Sejong-si, South Korea.

Koo, B., Wilson, G.M., Morris, R.E., Dunker, A.M., Yarwood,
G. (2009) Comparison of source apportionment and sensitivity analysis in a particulate matter air quality model. Environmental Science and Technology 43(17), 6669–6675.
Kwon, T.H., Park, M.-S., Yi, C., Choi, Y.J. (2014) Effects of different averaging operators on the urban turbulent fluxes. Atmosphere Korean Meteorological Society 24, 197–206, DOI: 10.14191/Atmos.2014.24.2.197.
Li, X., Wang, Y., Zhao, H., Hong, Y., Liu, N., Ma, J. (2018) Characteristics of pollutants and boundary layer structure during two haze events in summer and autumn 2014 in Shenyang, Northeast China. Aerosol and Air Quality Research 18, 386–396, DOI: 10.4209/aaqr.2017.03.0100.
NIER (National Institute of Environmental Research in Korea) (2017) Introduction to the KORUS-AQ Rapid Science Synthesis Report, Incheon, South Korea, 24 pp.
Park, I.-S. (2014) Creative counter-measures of PM$_{10}$. Journal of Korean Society for Atmospheric Environment 30(2), 211–212.
Park, I.-S. (2016) Why do the high concentration PM$_{10}$ episodes occur and what is the major cause of the episodes? Journal of Korean Society for Atmospheric Environment 32(3), 352–353.
Park, I.-S., Choi, W.-J., Lee, T.-Y., Lee, S.-J., Han, J.-S., Kim, C.-H. (2005) Simulation of long-range transport of air pollutants over Northeast Asia using a comprehensive acid deposition model. Atmospheric Environment 39, 4075–4085, DOI: 10.1016/j.atmosenv.2005.03.038.
Park, I.-S., Lee, S.-J., Kim, C.-H., Yoo, C., Lee, Y.-H. (2004) Simulating urban-scale air pollutants and their predicting capabilities over the Seoul Metropolitan Area. Journal of the Air & Waste Management Association 54, 695–710.
Park, I.-S., Song, C.-K., Park, M.-S., Kim, B.-G., Jang, Y.-W., Ha, S.-S., Jang, S.-H., Chung, K.-W., Lee, H.-J., Lee, U.-J., Kim, S.-K., Kim, C.-H. (2018) Numerical study on the impact of power plants on primary PM$_{10}$ concentrations in South Korea. Asian Journal for Atmospheric Environment 12(3), 255–273, DOI: 10.5572/ajae.2018.12.3.255.
Park, M.-S. (2018) Overview of meteorological surface variables and boundary-layer Structures in the Seoul Metropolitan Area during the MAPS-Seoul Campaign. Aerosol and Air Quality Research 18, 2157-2172. DOI: 10.4209/aaqr.2017.10.0428.
Park, M.-S., Chae, J.-H. (2018) Features of sea-land-breeze circulation over the Seoul Metropolitan Area. Geoscience Letters 5, 28, DOI: 10.1186/s40562-018-0127-6.
Park, M.-S., Joo, S.J., Park, S.-U. (2014) Carbon dioxide concentration and flux in an urban residential area in Seoul, Korea. Advances in Atmospheric Sciences 31, 1101–1112, DOI: 10.1007/s00376-013-3168-y.
Park, M.-S., Park, S.-H., Chae, J.-H., Choi, M.-H., Song, Y., Kang, M., Rho, J.-W. (2017) High-resolution urban observation network for user-specific meteorological information service in the Seoul Metropolitan Area, South Korea. Atmospheric Measurement Techniques 10, 1575–1594, DOI: 10.5194/amt-10-1575-2017.
Park, S.-U., Lee, I.H., Choe, A., Joo, S.J. (2015) Contributions of the pollutant emission in South Korea to the aerosol concentrations and depositions in Asia. Asia-Pacific Journal of Atmospheric Sciences 51, 183–195, DOI: 10.1007/s13143-015-0069-2.
Seo, J., Kim, J.Y., Yoon, D., Lee, J.Y., Kim, H., Lim, Y.B., Kim, Y., Jin, H.C. (2017) On the multiday haze in the Asian continental outflow: the important role of synoptic conditions combined with regional and local sources. Atmospheric Chemistry and Physics 17, 9311–9332, DOI: 10.5194/acp-17-9311-2017.
Seo, J., Park, D.-S. R., Kim, J.Y., Youn, D., Lim, Y.B., Kim, Y. (2018) Effects of meteorology and emissions on urban air quality: a quantitative statistical approach to long-term records (1999–2016) in Seoul, South Korea. Atmospheric Chemistry and Physics 18, 16121–16137, 2018, DOI: 10.5194/acp-18-16121-2018.
Venkatram, A., Karamchandani, P., Pai, P., Sloane, C., Saxena, P., Goldstein, R. (1997) The development of a model to examine source-receptor relationships for visibility on the Colorado Plateau. Journal of the Air and Waste Management Association 47, 286-301, DOI: 10.1080/10473289.1997.1046453.
Wang, S., Zhou, P., Lin, L., Liu, C., Huang, T. (2018) Influence of major urban construction on atmospheric particulates and emission reduction measures. Asian Journal for Atmospheric Environment 12(3), 215-231, DOI: 10.5572/ajae.2018.12.3.215.