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Temporal and Spatial Variations in Fine and Coarse Particles in Seoul, Korea

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ABSTRACT

Concentrations of fine ($PM_{2.5}$) and coarse ($PM_{10–2.5}$) particles, whose aerodynamic diameters are less than or equal to 2.5 µm, and greater than 2.5 and less than or equal to 10 µm, respectively, at ambient air monitoring stations in Seoul between 2002 and 2008 were analyzed. Effects of Asian dust are mainly manifested as concentration spikes of $PM_{10–2.5}$, but were considerable on $PM_{2.5}$ levels in 2002 when Asian dust storms were the strongest. Excluding the effects of Asian dust, annual average $PM_{2.5}$ showed a downward trend. Despite a similarity in year-to-year variations, $PM_{10–2.5}$, mostly affected by fugitive dust emissions, and CO and NO_2, primarily affected by motor vehicle emissions, did not show a decrease. $PM_{2.5}$ along with CO and NO_2 had peak concentration during the morning rush hour; the $PM_{10–2.5}$ peak lagged one hour behind the $PM_{2.5}$ peak. On high $PM_{2.5}$ days, $PM_{2.5}$ peaks occurred two hours later than usual as the effects of secondary formation through photochemical reactions became more important. A test for the spatial variability shows that $PM_{10–2.5}$, which is known to be greatly influenced by local effects, is lower in its correlation coefficient and higher in its coefficient of divergence (COD, which serves as an indicator for spatial variability) than $PM_{2.5}$, albeit by only a small difference. The average COD of $PM_{2.5}$ among monitoring stations was about 0.2 but was lowered to 0.13 when considering high $PM_{2.5}$ days only, signifying that spatial uniformity increases due to the pervasive influence of photochemical reactions.

Keywords: Time trends; Spatial variability; Fugitive dust; Vehicular emissions; High $PM_{2.5}$ days.

INTRODUCTION

Particulate matter (PM) is usually considered a single entity. However, PM consists of various constituents that can be grouped into inorganic ions, carbonaceous materials, and trace elements. In the eastern United States and Western Europe, inorganic ions and carbonaceous materials are major constituents of PM (NARSTO, 2004; Putaud *et al.*, 2004). Around the dust belt extending from the west coast of North Africa, over the Middle East, to China (Prospero *et al.*, 2002), the portion of crustal materials, mainly composed of inorganic ions and oxides of trace elements, is larger (Arimoto *et al.*, 2006; Koçak *et al.*, 2007; Yuan *et al.*, 2008). Outside the dust belt, the effects of forest fires and biomass burning, including the burning of agricultural or crop waste, are significant (Duan *et al.*, 2004; Giglio *et al.*, 2006; Kim *et al.*, 2007). Korea is located downwind of the prevailing westerlies in the easternmost area of the Asian continent (Fig. 1), and thus is significantly influenced by biomass burning and fugitive dust represented by Asian dust.

In Korea, PM_{10} was designated as a criteria pollutant in 1995. Annual averages of PM_{10} in Seoul have decreased continuously from 78 µg/m^3 in 1995 to 55 µg/m^3 in 2008 with year-to-year fluctuations (NIER, 2012). Nevertheless, the level of PM_{10} in Seoul is much higher than those in New York and Paris, which vary around 20 µg/m^3, and that in Tokyo, which has recently been reduced to the 20s µg/m^3 (KOSAE, 2009). It has been pointed out that fugitive dust is the primary cause of higher PM_{10} concentrations in Seoul. Park *et al.* (2010) indicated that, through regional-scale air quality modeling, annual-average PM_{10} concentrations over the Korean peninsula in 2007 can increase by about 25 µg/m^3 because of fugitive dust emissions in northeast Asia. Chemical mass balance receptor modeling based on 2006–2007 monitoring data in Seoul showed that fugitive dust accounts for about 25–31% of total PM_{10}, following secondary formation of about 33–48% (KOSAE, 2009).

In Korea, ambient air quality standards for $PM_{2.5}$ were established in 2011 and will become effective in 2015. To date, PM research using air monitoring network data in Korea has targeted for PM_{10} (Han *et al.*, 2008; Kim and Shon, 2011; Choi *et al.*, 2013), whereas research for $PM_{2.5}$ was focused on monitoring data at a specific location (Kang *et al.*, 2004; Park *et al.*, 2007; Heo *et al.*, 2009). In

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this study, $PM_{2.5}$ concentration data from the air monitoring network are examined, together with PM_{10} concentration data. In particular, along with the distribution of buses powered with compressed natural gas (CNG) since the 2000s (Kang, 2004; Nguyen *et al.*, 2010), the "Special Act on Seoul Metropolitan Air Quality Improvement" came into effect in 2005, leading to emissions reduction from diesel vehicles and fugitive dust emissions on roads and open areas (KOSAE, 2009). Thus, this study is meaningful in evaluating the changes in PM concentration levels and in tracking the effectiveness of emissions reduction strategies.

DATA AND METHODS

One-hour averages of PM_{10} and $PM_{2.5}$ were measured using the beta-ray absorption method after fractionation of suspended particles with an impactor (KME and NIER, 2009). There were 27 stations in Seoul performing urban air quality monitoring between 2002 and 2008, and PM_{10} was measured at all of them. $PM_{2.5}$ has also been measured to establish the basis for the designation of a criteria pollutant. The number of stations to measure $PM_{2.5}$ has been increasing. In this study, concentrations of PM_{10} and $PM_{2.5}$ from 14 stations, where both were continuously measured during the study period, are analyzed. Concentration of $PM_{10-2.5}$ (coarse particles) was obtained by subtracting $PM_{2.5}$ (fine particles) from PM_{10}. Monthly and annual averages were calculated from 1-h averages when at least 75% of the data are available, which is the criterion for determining the average.

Fig. 1 shows the locations of 14 stations whose data were used in this work. An area-wide mean of annual averages for Seoul was obtained from annual averages from the stations, whereas the official annual mean reported by the Korean Ministry of Environment is calculated with 1-h averages from all stations. The differences between the two means were negligible for PM_{10} because the data recovery rates at most stations were much higher than 75%. The area-wide PM_{10} means for Seoul obtained from 14 stations (63.7 ± 8.2 µg/m^3, which are represented as mean ± standard deviation) were slightly higher than those from all stations (62.9 ± 7.3 µg/m^3) during the study period, but the difference was not statistically significant (*t*-test, $p > 0.05$).

To facilitate the understanding of variations in PM concentrations, concentrations of gaseous criteria pollutants such as CO, NO_2, and ozone were also analyzed. One-hour averages for these criteria pollutants are measured at all 14 monitoring stations in Fig. 1 using nondispersive infrared, chemiluminescent, and ultraviolet photometric methods, respectively (KME and NIER, 2009). Meteorological data from the Seoul Weather Station were used in this study (see Fig. 1 for the location).

RESULTS AND DISCUSSION

Annual Averages

Figs. 2(a) and 2(b) show the variations in annual averages of $PM_{2.5}$ and $PM_{10-2.5}$ for all days and excluding Asian dust (AD) days, respectively. Here, the AD day denotes the day for which the Korea Meteorological Administration officially announced that AD was observed. Annual average denotes the area-wide mean of annual averages for Seoul. Annual averages of both fine and coarse particles are the highest in 2002 for all days but are highest in 2003 if excluding AD days. Because the number of AD days and the 24-h PM on the AD days were the highest in 2002 (Fig. 2(c)), the highest annual averages of fine and coarse particles for that year in Fig. 2(a) were attributable to the effects of AD. Note that AD particles are mainly composed of coarse mineral particles. This outcome is corroborated by the lower $PM_{2.5}/PM_{10}$ ratio (PM ratio), indicating a larger fraction of coarse particles (Fig. 2(a)) and by a decline of both concentrations in 2002, excluding AD days (Fig. 2(b)). The 24-h PM on the AD days rises again in 2006. However, the increase in PM concentrations accompanied with the reduction of the PM ratio occurs in 2007. PM concentrations in 2007 are still high in Fig. 2(b), excluding AD days. An increase in PM concentrations in 2007 presumably occurred as a result of other factors (e.g., local fugitive dust emissions or meteorology), rather than the effects of AD.

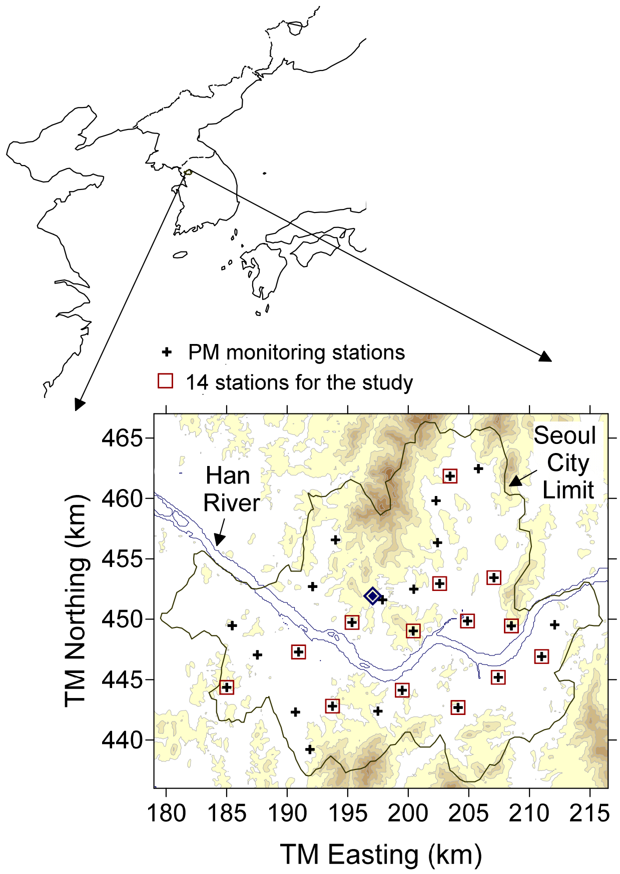

Fig. 1. Location of 14 monitoring stations continuously measuring both PM_{10} and $PM_{2.5}$ during the 2002–2008 study period among all stations in Seoul. Diamond denotes the Seoul Weather Station. TM stands for Transverse Mercator. Topographic features are also shown with the Han River running westward through the city.

During the study period, both $PM_{2.5}$ and the PM ratio generally decrease regardless of inclusion of the AD days. As mentioned, the Korean government has undertaken

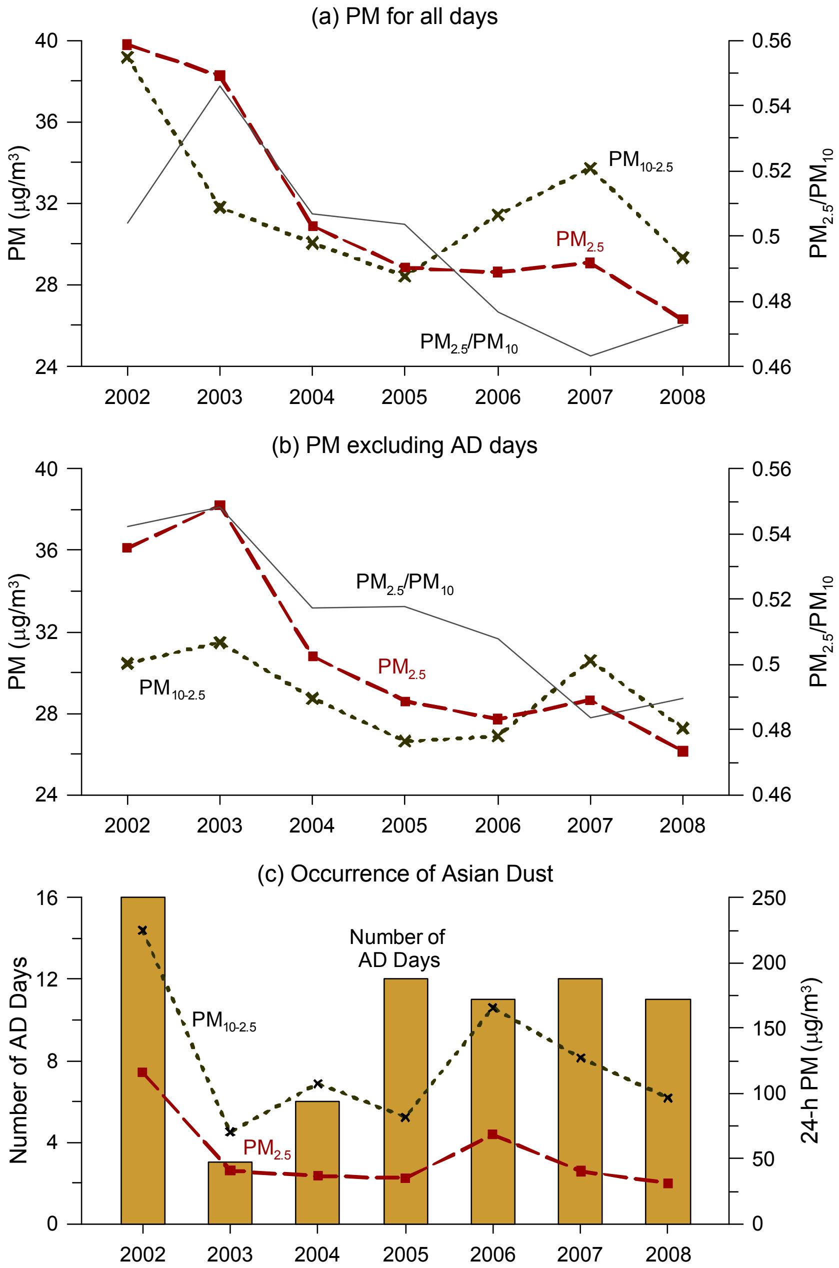

Fig. 2. Trends in annual averages of fine ($PM_{2.5}$) and coarse particles ($PM_{10–2.5}$): (a) for all days, and (b) excluding Asian dust (AD) days. Trends in the annual number of AD days along with mean 24-h PM concentrations on AD days are shown in (c).

extensive and costly efforts during the past several years to reduce particle pollution in the greater Seoul area, including in Seoul proper and its neighboring satellite cities. Although the government's measures address various emissions of PM and its precursor gases, considerable effort has been devoted to the control of particle emissions from diesel vehicles. Furthermore, anthropogenic emissions of PM and its precursor gases associated with rapid economic growth in China have resulted in a substantial increase throughout the 2000s (Zhang *et al.*, 2009). It is certain that efforts by the Korean government have some effects although the effectiveness of each effort has not yet been fully assessed.

In contrast to $PM_{2.5}$, the trend in $PM_{10–2.5}$ variation is unclear. In Fig. 2(a), a decrease in $PM_{10–2.5}$ can be seen in subsequent years after the high concentrations measured in 2002. These variations in $PM_{2.5}$ and $PM_{10–2.5}$ are similar to those reported by Minoura *et al.* (2006) who investigated the PM trend in Tokyo to evaluate the effects of emission reduction measures for motor vehicles. During 1994–2004, suspended particulate matter (~PM_7) decreased from 52 μg/m^3 to 31 μg/m^3. Most of the decrease was attributable to the reduction of fine particles (~$PM_{2.1}$) while the annual average of coarse particles (~$PM_{7–2.1}$) varied around the 10–15 μg/m^3 range. If comparing PM in Fig. 2(b) excluding AD days with those in Tokyo, $PM_{2.5}$ of 26 μg/m^3 in 2008 is still higher than $PM_{2.1}$ of below 20 μg/m^3 in Tokyo in 2004; and further, $PM_{10–2.5}$ of around 29 μg/m^3 between 2002–2008 is much higher than $PM_{7–2.1}$ in Tokyo during 1994–2004. One of the reasons for the discrepancies between the two studies is the different size ranges of PM they considered. Nevertheless, it is apparent that more attention should be given to reducing the level of $PM_{10-2.5}$ as well as $PM_{2.5}$ in Seoul.

Variations in annual averages of gaseous pollutants, visual range, and meteorological variables are provided in Fig. 3 to assist in understanding the year-to-year variations in Figs. 2(a) and 2b). As mentioned previously, the highest PM_{10} occurs in 2002 because of the effects of AD. However, Fig. 3 shows that CO is the highest in 2007 followed by a peak in 2002. Furthermore, NO_2 is high in 2007 in which $PM_{2.5}$ and $PM_{10-2.5}$ also rise, as shown in Figs. 2(a) and 2(b); NO_2 is the highest in 2003 when $PM_{2.5}$ reaches peak values in Fig. 2(b). In the early 2000s, high concentrations of CO and NO_2 suggest that high concentrations of $PM_{2.5}$ are, to some extent, related to vehicle emissions. However, CO and NO_2 levels in 2007 are comparable to those in 2002 and 2003, whereas $PM_{2.5}$ in 2007 is much lower than those in 2002 and 2003. Steady visibility degradation (i.e., decreasing visual range) and increases in both mean and peak ozone are also different from the variation of $PM_{2.5}$. In general, significant photochemical reactions that form high ozone can also produce high concentrations of secondary particles (mostly $PM_{2.5}$). However, in this study, their effects are noticeable only on high $PM_{2.5}$ days, as is discussed later.

Annual variations in meteorological parameters are not much helpful in explaining the variations in PM in Figs. 2(a) and 2(b). Among meteorological parameters shown in

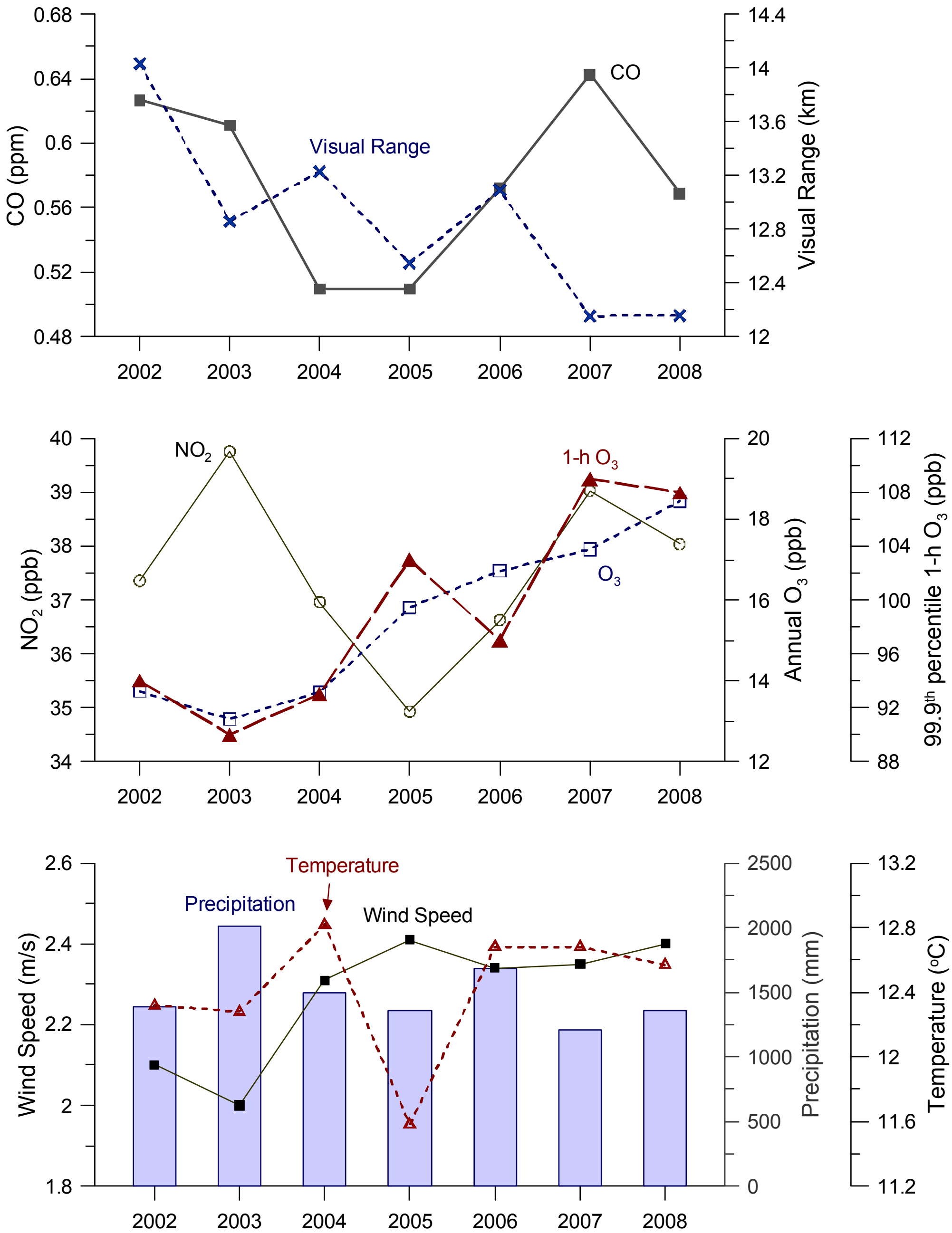

Fig. 3. Trends in annual averages of gaseous pollutants, visual range and meteorological variables.

Fig. 3, the variation in wind speed is most correlated with those in pollutants. Wind speed is the lowest in 2003 in which $PM_{2.5}$ in Fig. 2(b) and NO_2 is the highest. It is the highest in 2005 in which $PM_{10-2.5}$ is the lowest in Figs. 2(a) and (b). The coefficient of determination (r^2) of wind speed is the highest at 0.95 with $PM_{2.5}$ in Fig. 2(b). The next highest is 0.63 with $PM_{10-2.5}$ in Fig. 2(b), and 0.61 and 0.57 with mean and peak ozone in Fig. 3, respectively.

Monthly and Hourly Averages

Fig. 4 shows the variations in monthly averages of PM and other pollutants. In March and April, high $PM_{10-2.5}$ concentrations were observed, which were associated with AD events. This correlation can be ascertained by the reduction of $PM_{10-2.5}$ concentration if excluding AD days in Fig. 4(b). However, even excluding AD days, $PM_{10-2.5}$ concentrations in March and April are somewhat higher than those in other months. This finding is attributable to high wind speed and dry conditions that are conducive to the generation of fugitive dust (KMA, 2001). Ozone concentration is high in May and June in Fig. 4(c) because of warm temperatures, along with long-range transport of ozone and its precursors from the Asian continent. But $PM_{2.5}$ levels are not high in these months even excluding AD days. This result is plausible when contributions of secondary formation by photochemical reactions are not are the lowest in August because of frequent precipitation scavenging of PM and precursor gases and/or because of

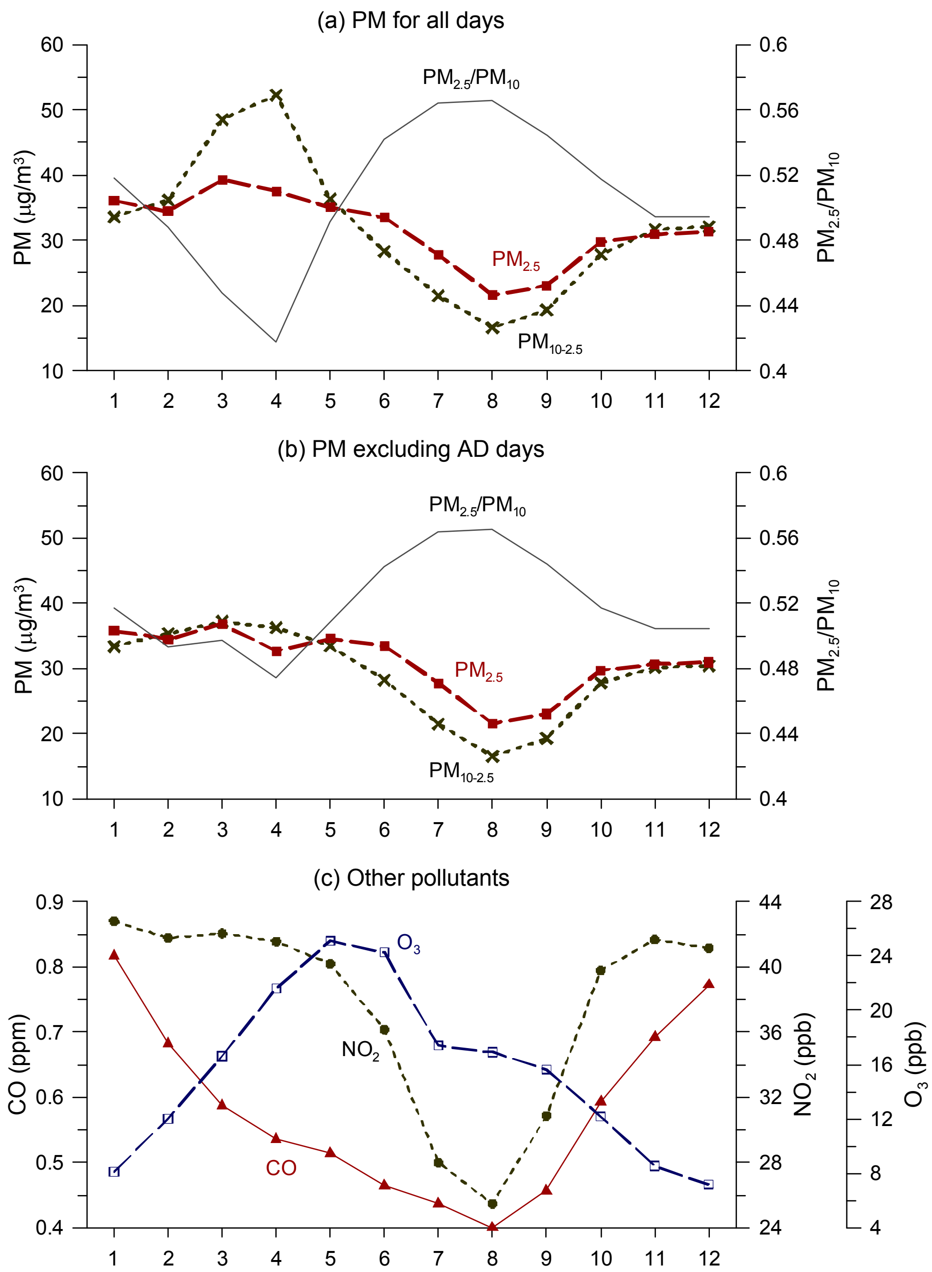

Fig. 4. Variations in monthly averages of: fine and coarse particles for (a) all days, (b) excluding Asian dust days; and (c) other pollutants.

large enough compared with those of primary emission from motor vehicles and fugitive dust. Both $PM_{10–2.5}$ and $PM_{2.5}$ inflow of clean air masses from the ocean into the Korean peninsula, as indicated in the variations in NO_2 and CO in Fig. 4(c). However, the PM ratio is the highest in August because precipitation can remove $PM_{10–2.5}$ more effectively than $PM_{2.5}$ (Seinfeld and Pandis, 1998).

Fig. 5 shows the variations in hourly averages of PM and other pollutants. PM concentrations are high in the morning and increase once again from the evening rush hour in Fig. 5(a). The variations in $PM_{10–2.5}$ and $PM_{2.5}$ are correlated but with some differences. First, $PM_{2.5}$ has a peak at 9 a.m., which is the average between 8 and 9 a.m., because of vehicle emissions during the morning rush hour. The peak concentrations of NO_2 and CO at the same time also support this hypothesis. However, the $PM_{10–2.5}$ peak occurs one hour later, at 10 a.m. Because the mixing height already begins to grow after sunrise, the $PM_{10–2.5}$ concentration cannot increase without additional emission and/or generation after the $PM_{2.5}$ peak. Moore *et al.* (2010) also observed the lag of peak concentration of $PM_{10–2.5}$ behind that of $PM_{2.5}$ in the morning in the Los Angeles area. It is construed that generation of paved road dust, consisting mostly of $PM_{10–2.5}$, plays a role as wind speed increases and relative humidity decreases despite increasing mixing height in the morning (Ghim *et al.*, 2005; Choi *et al.*, 2014). Thus, the $PM_{10–2.5}$ peak occurred while increasing mixing heights counteract increasing fugitive dust emissions.

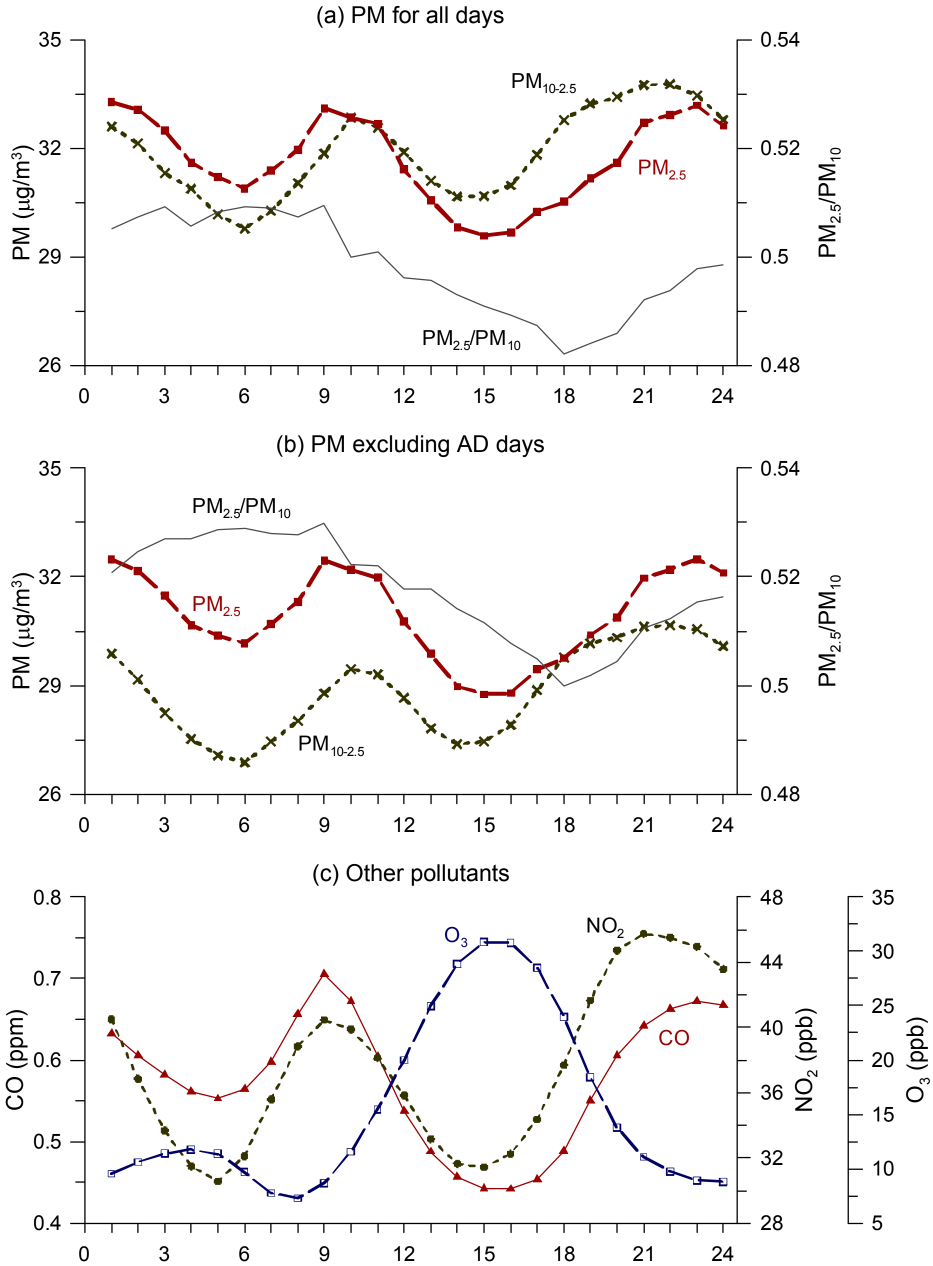

Fig. 5. Variations in hourly averages of: fine and coarse particles for (a) all days, (b) excluding Asian dust days; and (c) other pollutants.

Second, $PM_{10–2.5}$ concentration is higher than $PM_{2.5}$ in the afternoon while the reverse is mostly true in the morning (Fig. 5(a)). As a result, the PM ratio in the morning is higher than that in the afternoon. This result occurs because of an increase in fugitive dust emissions in the afternoon (Choi *et al.*, 2014). Excluding AD days, diurnal variations of $PM_{10–2.5}$, $PM_{2.5}$, and the PM ratio are similar except for the lowered $PM_{10–2.5}$ concentration and elevated PM ratio (Fig. 5(b)). Both CO and NO_2 can be tracers of vehicle emissions. Toward the evening, the variation in $PM_{2.5}$ is similar to that in CO, while the variation in $PM_{10–2.5}$ is similar to that in NO_2. As mentioned earlier, variations in $PM_{2.5}$ represent the effects of vehicle emissions, while being affected by diurnal variations in mixing height. Note that the NO_2 peak is higher in the late evening than in the morning (Fig. 5(c)), similar to $PM_{10–2.5}$. A higher peak of NO_2 in the late evening is due to its production from ozone titration by NO after sunset (David and Nair, 2011), being different from the $PM_{10–2.5}$ peak caused by the generation of paved road dust under a shallow mixing height.

High $PM_{2.5}$ Days

Up to now, many aspects of the variations in $PM_{10–2.5}$ and $PM_{2.5}$ were discussed in terms of vehicle emissions and fugitive dust, as well as the AD effects. Such an approach was particularly useful in interpreting the diurnal variations in PM in Fig. 5. However, it is unusual that the effect of secondary formation, which is common in the areas with similar conditions, was not observed (Wittig *et al.*, 2004; Fine *et al.*, 2008; Millstein *et al.*, 2008; Turner and Allen, 2008). To elucidate this difference, variations in PM on high $PM_{2.5}$ days are investigated in Fig. 6 when the contribution of secondary formation is generally high in many areas.

Here, high $PM_{2.5}$ denotes that 24-h $PM_{2.5}$ is greater than or equal to 56 μg/m^3, corresponding to the highest 10% excluding AD days. In comparison with annual variations Here, high $PM_{2.5}$ denotes that 24-h $PM_{2.5}$ is greater than or equal to 56 μg/m^3, corresponding to the highest 10% excluding AD days. In comparison with annual variations only excluding AD days in Fig. 2(b), there is no distinct trend in $PM_{2.5}$ in Fig. 6(a). Nevertheless, higher values can be seen in 2003 when annual averages of $PM_{2.5}$ and NO_2 were the highest in Figs. 2(b) and 3, respectively. Although the PM ratio also decreases, in this case, it is primarily due to the increase in $PM_{10–2.5}$ after 2005.

As shown in Fig. 6(b) for high $PM_{2.5}$ days, monthly $PM_{2.5}$ is highest in September, and lower concentrations in July through September, which are typical for all days in Fig. 4(b), are hardly discernible. In summer in Seoul, (1) high temperatures around 30°C are favorable for photochemical reactions; (2) westerly winds that bring pollutants from the Asian continent over the Korean Peninsula are least common; and (3) scavenging by frequent precipitation inhibits the buildup of pollutants (Ghim *et al.*, 2001; KMA, 2001). Despite (1), summertime $PM_{2.5}$ concentrations are lower than in other seasons because of the effects of (2) and (3), as shown in Fig. 4(b). However, $PM_{2.5}$ averages on high $PM_{2.5}$ days in July and August are similar to other months and higher in September, being predominated by effects of (1).

On high $PM_{2.5}$ days, peak $PM_{2.5}$ occurs at 11 a.m., which lags two hours, compared with the result from excluding AD days shown in Fig. 5(b). Millstein *et al.* (2008) observed peak concentration of $PM_{2.5}$ nitrate between 8 a.m. and noon at four sites in the United States, and earlier in the day in spring and summer compared to fall and winter. They explained that this result occurred because of a combination of factors including ammonium nitrate dissociation and the onset of convective mixing that occurs earlier in the day in spring and summer. Measurement with PILS (Particle-into-Liquid Sampler) at a site downwind of Seoul also showed that concentrations of major secondary ions such as sulfate and nitrate were the highest at 11 a.m. concurrently with high $PM_{2.5}$ concentration (Lee *et al.*, 2012).

Fig. 7 shows the variations in hourly averages of gaseous pollutants on high $PM_{2.5}$ days. Both NO_2 and CO peaks occurred at 9 a.m. for all days (Fig. 5(b)); however, the NO_2 peak occurs at 10 a.m., one hour later than the CO peak in Fig. 7. It is common that the NO_2 peak occurs between the NO and O_3 peaks (Fujita *et al.*, 2003). This result occurs because NO is a primary pollutant and O_3, a secondary pollutant, is produced with decreasing NO_2 in a typical photochemical reaction system. Similarly, it can be reasonably anticipated that nitrate exhibits a peak after the NO_2 peak because nitrate is formed from NO_2 (Seinfeld and Pandis, 1998). An analogous behavior for sulfate can be postulated. However, it seems that their peaks did not occur later than those shown in Fig. 6(c), at least because of the onset of convective mixing with increasing temperature, as indicated by Millstein *et al.* (2008).

Spatial Variabilities

It is well known that spatial distribution of PM_{10} is more variable than that of $PM_{2.5}$ (NARSTO, 2004) because of a shorter residence time and more sporadic sources of $PM_{10–2.5}$ in comparison with $PM_{2.5}$. Selected statistics showing the spatial variabilities of fine and coarse particles for all days and high $PM_{2.5}$ days are given in Table 1. These statistics were calculated from 24-h averages to compare their values with the previous studies and to examine the variability of concentration levels excluding the diurnal variations (Pinto *et al.*, 2004; Wilson *et al.*, 2005; Moore *et al.*, 2010). The mean, standard deviation, maximum, and minimum are obtained from the values of 91 possible combinations of two stations chosen from 14 stations (i.e., ${}_{14}C_2$). The correlation coefficient provides a measure of similarity in temporal variations. On the other hand, the 90th percentile (P_{90}) of the absolute difference between the two stations provides an absolute measure of the spatial variability, and the coefficient of divergence (COD) provides a relative measure of the variability across stations. The COD is defined by (Pinto *et al.*, 2004; Wilson *et al.*, 2005):

$$\mathrm{COD}_{jk} = \sqrt{\frac{1}{N}\sum_{n=1}^{N}\left(\frac{x_{nj}-x_{nk}}{x_{nj}+x_{nk}}\right)^2} \tag{1}$$

where x_{nj} and x_{nk} represent the 24-h concentration for day n

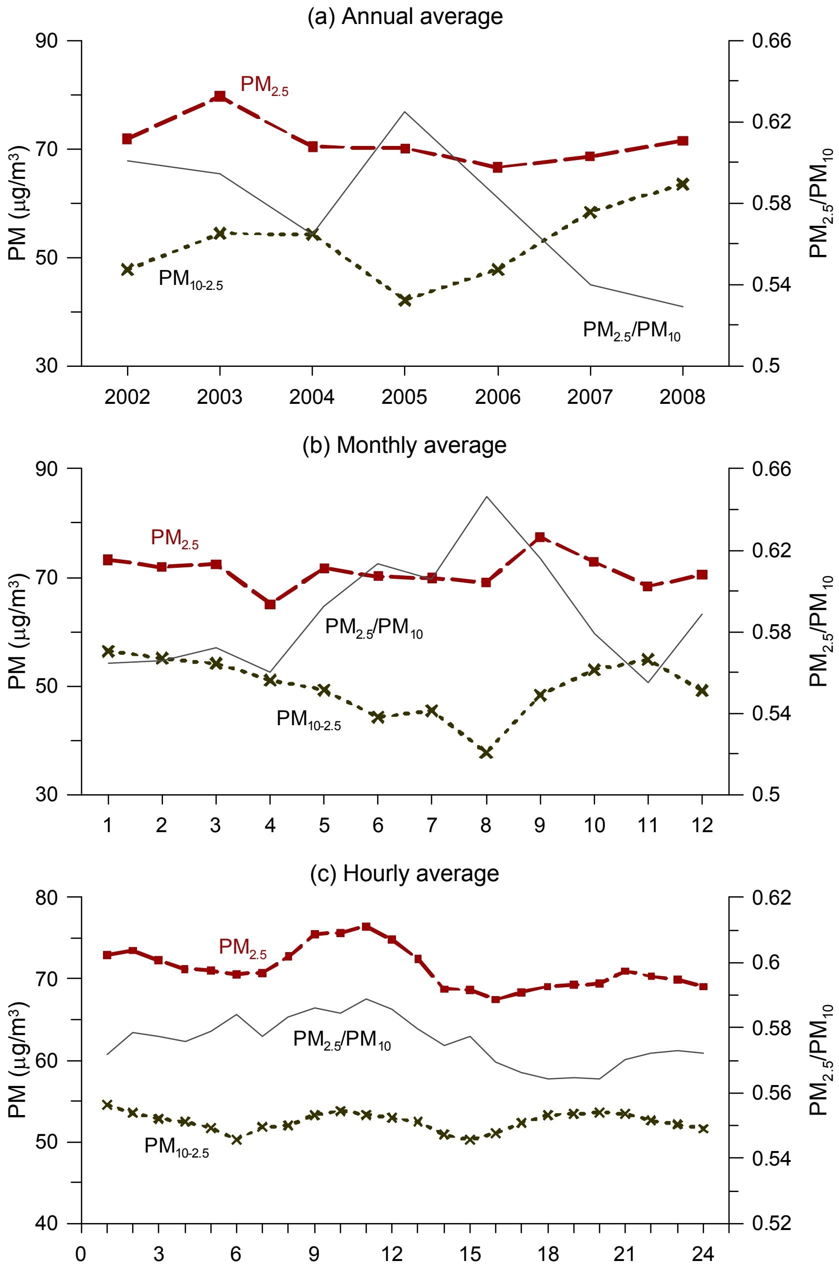

Fig. 6. Temporal variations in fine and coarse particles on high $PM_{2.5}$ days (excluding AD days).

at station *j* and *k*, and *N* is the number of observations. A COD of zero means that two data sets are exactly the same (spatial homogeneity), and a value approaching one indicates maximum difference or variability (extreme spatial heterogeneity).

In Table 1, the correlation coefficients are generally higher for $PM_{2.5}$ than those for $PM_{10-2.5}$ as mentioned at the beginning of this section. Although small, their difference is statistically significant (*t*-test, $p = 0.000$) as indicated by their smaller standard deviations. A small difference between $PM_{2.5}$ and $PM_{10-2.5}$ is probably because all sites are located within the same metropolitan area, similarly influenced by vehicles and fugitive dust, although local environmental settings, including topography, are quite different (Fig. 1). In fact, absolute values of the correlation coefficient of $PM_{10-2.5}$ between the sites in the Los Angeles area were 0.69–0.89 within urban areas but went down to 0.42 when the sites in semi-rural areas were included (Pakbin *et al.*, 2010). The mean value of P_{90} for $PM_{2.5}$ is slightly higher than that for $PM_{10-2.5}$, but they are not statistically different. The lower mean value of COD for $PM_{2.5}$ ($p = 0.000$) indicates that $PM_{2.5}$ is more uniform than $PM_{10-2.5}$, as seen in the correlation coefficient.

On high $PM_{2.5}$ days, differences between $PM_{2.5}$ and $PM_{10-2.5}$ are greater than those for all days and statistically significant ($p < 0.05$). In particular, a COD for $PM_{2.5}$ is reduced greatly because differences among monitoring stations diminish as high $PM_{2.5}$ days consider the upper 10th percentile of $PM_{2.5}$ only. As the range of variations in $PM_{2.5}$ and the number of data were reduced, correlation

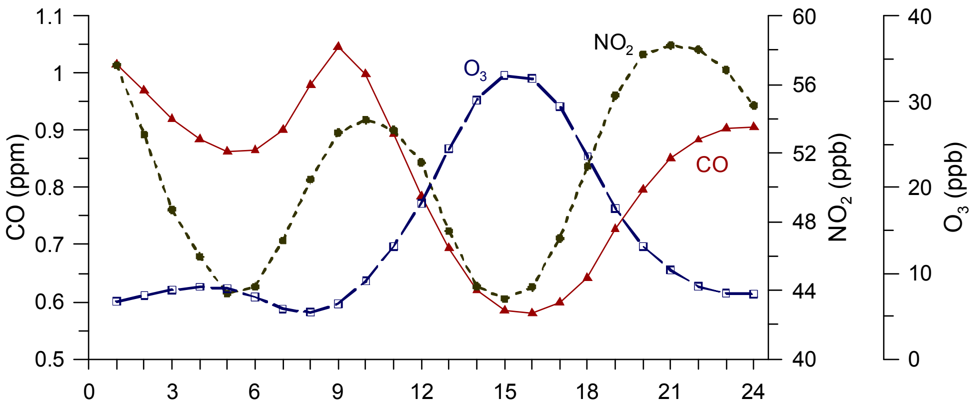

Fig. 7. Variations in hourly averages of gaseous pollutants on high $PM_{2.5}$ days (excluding AD days).

Table 1. Comparison of statistics showing the spatial variabilities of fine and coarse particle concentrations between all days and high $PM_{2.5}$ days.[a]

		Correlations[b]	P_{90} ($\mu g/m^3$)[c]	COD[d]
All days	$PM_{2.5}$	0.82 ± 0.05 (0.62–0.92)[e]	21.1 ± 5.5 (11.0–33.7)	0.20 ± 0.04 (0.10–0.31)
	$PM_{10-2.5}$	0.74 ± 0.06 (0.56–0.89)	20.7 ± 2.7 (14.0–27.1)	0.22 ± 0.03 (0.16–0.27)
High $PM_{2.5}$ days	$PM_{2.5}$	0.56 ± 0.13 (0.15–0.81)	36.4 ± 10.2 (18.7–58.0)	0.12 ± 0.03 (0.08–0.19)
	$PM_{10-2.5}$	0.44 ± 0.16 (–0.01–0.74)	38.9 ± 7.6 (25.5–61.1)	0.21 ± 0.03 (0.14–0.29)

[a] Excluding AD days in order to estimate the characteristics of high $PM_{2.5}$ days in comparison with normal days. This approach was because high $PM_{2.5}$ days are those on which 24-h average $PM_{2.5}$ are included in the highest 10% excluding AD days.
[b] Pearson correlation coefficient.
[c] 90th percentile of the absolute values of the difference.
[d] Coefficient of divergence.
[e] Mean ± standard deviation (maximum–minimum).

coefficients among monitoring stations become lower, which is similar for $PM_{10-2.5}$. P_{90} becomes larger because differences among monitoring stations become larger on high $PM_{2.5}$ days while the number of data is reduced.

Although uniformity of spatial variations is determined in the relative sense, a COD of 0.2 has often been used as the criterion for determining uniformity (Wilson *et al.*, 2005). Only considering COD with the same criterion, $PM_{2.5}$ on high $PM_{2.5}$ days can be judged to be uniform. However, as mentioned previously, temporal variations among monitoring stations showed low correlations despite uniformity of spatial variations. Pinto *et al.* (2004) examined the items in Table 1 by classifying the $PM_{2.5}$ measurement data of the consolidated metropolitan statistical area (CMSA) designated by the U.S. Environmental Protection Agency into East, Midwest, and West. Seoul is close to the West because correlation coefficients of $PM_{2.5}$ are not high and CODs on all days are not low (Table 1). Annual average $PM_{2.5}$ of 30 $\mu g/m^3$ and higher P_{90} in Table 1 are also similar to Los Angeles among U.S. CMSAs.

SUMMARY AND CONCLUSIONS

Temporal and spatial variations in $PM_{2.5}$ and $PM_{10-2.5}$ are investigated based on monitoring data of 14 stations where both PM_{10} and $PM_{2.5}$ have been measured continuously during the 2002–2008 period. Temporal variations are examined based on averages from 14 stations in Seoul. Overall, annual average $PM_{10-2.5}$ is strongly influenced by Asian dust in terms of annual averages but effects of Asian dust on $PM_{2.5}$ were most pronounced in 2002 when the Asian dust storms were severe. Excluding Asian dust days, annual average $PM_{2.5}$ shows a clear downward trend. However, $PM_{10-2.5}$, strongly affected by fugitive dust, and CO and NO_2, greatly affected by motor vehicle emissions, do not decrease despite their similarity in year-to-year variations. These variations exhibit relatively high correlation with wind speed among meteorological parameters, which can explain year-to-year variations but not pronounced enough to account for trends over the whole period.

The effects of Asian dust on both $PM_{10-2.5}$ and $PM_{2.5}$ have been observed in April and May when the Asian dust is most frequent. Even excluding Asian dust days, weather conditions are conducive to fugitive dust generation in those months, and thus $PM_{10-2.5}$ levels are higher than other months. Because of scavenging effects by frequent precipitation in July through September, concentration levels of both $PM_{10-2.5}$ and $PM_{2.5}$ were lowered but particularly for $PM_{10-2.5}$. However, monthly averages on high $PM_{2.5}$ days, which include upper 10^{th} percentile $PM_{2.5}$ concentrations only, are not lower in July and August and highest in September because of secondary formation through photochemical reactions.

During the morning rush hour, $PM_{2.5}$ peaked along with

NO_2 and CO because of vehicular emissions. However, $PM_{10-2.5}$, greatly influenced by fugitive dust emissions, exhibited a maximum around 10 a.m., one hour after the morning rush hour. Both $PM_{2.5}$ and $PM_{10-2.5}$ also exhibited another peak in the late evening along with CO and NO_2. $PM_{10-2.5}$ peak in the late evening is larger than that in the morning due to the effects of fugitive dust emissions with a lowered mixing height, while NO_2 varies similarly but its evening peak is higher than its morning peak because of the production from ozone titration by NO.

On high $PM_{2.5}$ days, $PM_{2.5}$ and NO_2 peak around 11 a.m. and around 10 a.m., respectively, which are two hours and one hour later than when considering all days. Toward the afternoon, secondary formation becomes intense (which is not pronounced on non-high $PM_{2.5}$ days) and the mixing height grows (which prevents accumulation of air pollutants). Thus, $PM_{2.5}$ tends to be the highest in late morning when combined effects of primary emissions, secondary formation, and the variation in mixing height are optimal.

$PM_{10-2.5}$, known to be greatly affected by local effects, shows a lower correlation coefficient and larger COD than $PM_{2.5}$. Despite differences in local environmental settings including topographic features, there is little difference because emission patterns represented by vehicular emissions and fugitive dust generation are similar within a relatively small metropolitan area (about 605 km^2). The COD of $PM_{2.5}$ and P_{90} in Seoul is somewhat higher than those in major U.S. cities but is comparable to those in Los Angeles. However, considering high $PM_{2.5}$ days only, spatial uniformity significantly increases with a COD of 0.13, while the correlation coefficients decrease because a small number of high $PM_{2.5}$ days are taken into account.

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