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Research Article

Studying Air Pollutants Origin and Associated Meteorological Parameters over Seoul from 2000 to 2009

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We investigate the temporal characteristics of major air pollutants collected from 44 air quality stations over the city of Seoul, Korea, namely, nitrogen dioxide, carbon monoxide, particular matter at 10 microns, and sulfur dioxide (SO_2) between 2000 and 2009. The corresponding satellite datasets, namely, aerosol optical depth (AOD_{sat}), Ångström exponent, and fine mode fraction, collected from moderate resolution imaging spectroradiometer (MODIS) as well as the Aeronet ground aerosol optical depth ($AOD_{aeronet}$), have been analyzed. Pollutants' seasonal effect has been inferred from the precipitation and temperature. The four pollutants under study show varying temporal characteristics with different annual mean concentration patterns. The monthly mean of mentioned pollutants all show similar low concentrations during the summer season and high concentrations during the winter season. We found that pollution is strongly linked to temperature and precipitation variability, especially during the fall season. Satellite data analysis provides information on the pollutants origin whether of natural or anthropogenic type. Our results indicate that the anthropogenic aerosol is dominant in the summer season even though the concentration was lower than the other seasons. $AOD_{aeronet}$ and Ångström exponent indicated high positive and negative correlation coefficients with PM_{10}, 0.60, and −0.45, respectively. Both small and large sizes of aerosols existed in 2007; however coarse size of aerosols was the primary component in 2002.

1. Introduction

Over the last few decades we have witnessed a large increase in the amounts and types of pollutants that emitted into the atmosphere since the Industrial Revolution. Their direct and indirect influences have been of concern due to their significant harm to human health, crops, and vegetation on regional and local (microzone) scales [1–4]. Although aerosols impact the global climate via different physical processes, yet a stronger impact on regional scales is particularly expected [5]. However, the physical characteristics, spatial distribution, and dynamics of aerosols are not completely understood [6–9]. Ground-based data enable the climatology of the aerosol attributes to be observed [10, 11], which can be verified by the application of satellite data [12, 13]. The origin of aerosols' effects and relationships with meteorological factors are essential elements in the study of mesoscale modeling, as well as regional impacts [14]. They can be of either natural origin such as dust storms, forest fires, and volcanoes or anthropogenic one such as electricity generation, traffic,

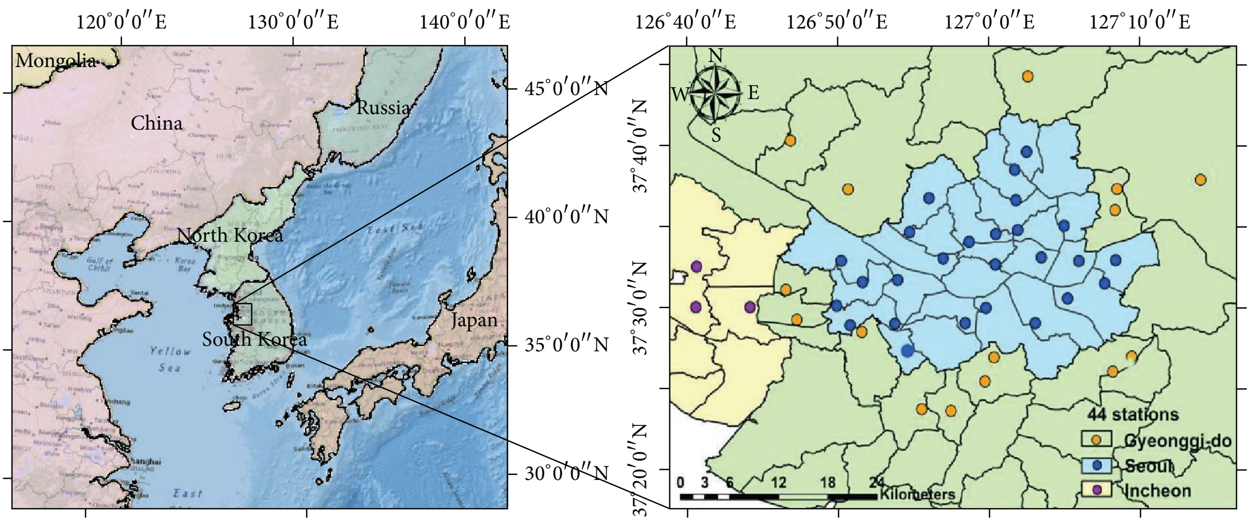

Figure 1: Base map of the study area, showing 44 air pollution monitoring stations.

wood fire heating, and industrial manufacturing processes [15–24].

East Asia has been experiencing frequent extremely high concentrations of sulfates, black carbon, nitrates, and organic matters due to increasing fossil fuel burning, as reported from the ACE-Asia campaign [25]. Previous work showed that the anthropogenic aerosols over the Eastern Asia were of much higher concentrations than those over Europe and the Eastern US [26, 27]. Kim et al. [28] investigated diurnal behavior and exceedance patterns of air quality criteria, ozone (O_3), and nitrogen dioxide (NO_2) in Seoul and showed that their behavior is strongly linked with geographical and meteorological factors. Harmful pollutants were observed at regional and temporal scales for particulate matter (PM_{10}) concentration over seven cities in Korea during the 2002 dust event as compared to the 2001 one [18].

Of particular concern is urban aerosol pollution, which has been the subject of many studies. Typical results of industrialization and urbanization have indicated many problems arising from urban aerosols [17, 29–33].

A great deal of research on aerosols has been carried out for several specific air pollutants in terms of their origin and relationship with meteorological parameters or seasonality using either ground or satellite observations [16, 34–38]. Using satellite observation data is an efficient way to determine and distinguish the optical properties of aerosols as it provides more complete coverage over longer time scales [32, 39–42]. Ground-based and satellite sensing of air pollutants may show different, but complementary characteristics; however, both are important in different situations, as well as for cross validation of the pollution origin [8].

Chang and Lee [34] investigated the temporal variation of air quality over Taipei city and found that primary pollutants such as carbon monoxide (CO), NO_x, and sulfur dioxide (SO_2) showed low concentration from local source activities when wind speed increases and the temperature drops. However, dusty days in Kuwait contributed to an evident decrease in temperature [43]. Choi et al. [44] found a possible relationship between cloud formation and PM_{10} on weekly timescales when studying the interaction between PM_{10} and the meteorological parameters in the boundary layer over China. Lee et al. [45] found that the annual mean SO_2 concentration in Seoul was higher than in Hamilton, Canada, and lower than in Chicago yet with the highest values during the winter season in Seoul, which is consistent with the high winter-time fuel usage.

The main purpose of the present work is to study the sources and origin of aerosol loading, being natural or anthropogenic, over Seoul, using ground and satellite observations between 2000 and 2009. We also investigate the relationships between pollutants, namely, NO_2, CO, SO_2, and PM_{10}, and meteorological parameters such as temperature, precipitation, and wind speed to determine the influence of meteorological conditions on pollutants concentrations.

2. Materials

2.1. Study Area. The study area is Seoul, the capital of South Korea, and its basin area, located in 126°62′E, 37°99′N and enclosed by mountains to the north and east (**Figure 1**). Seoul is characterized by noticeable distinction between the four seasons with warm weather in spring, hot and humid in summer, cool in fall, and cold and dry in winter [46].

Seoul has been experiencing serious pollution problems from local and external sources and in turns systemically polluting surrounding cities. In addition, it is one of the most densely populated cities in the world, with approximately 49% of Korea's entire population and 46% of vehicles holdings; however, it only occupies 12% of Korea's land area (605.25 km^2). Most NO_x and CO are emitted from vehicles, while SO_x and PM_{10} are emitted from manufacturing related combustion [47, 48]. Moreover, dust storms from China [49] also regularly cause an increase in the PM_{10} concentration and threaten people's health with respiratory and cardiovascular diseases [50].

Table 1: The characteristics and source of data used in this study.

Parameter	Time period	Frequency	Source
SO_2	2000–2009	Hourly	National Institute of Environmental Research (NIER)
NO_2	2000–2009	Hourly	
CO	2000–2009	Hourly	
PM_{10}	2000–2009	Hourly	
Dust frequency	2000–2009	Monthly	
Precipitation	2000–2007	Daily	Automatic Weather System (AWS)
Temperature	2000–2007	Daily	
Relative humidity (RH)	2000–2009	Monthly	Korea Meteorological Administration (KMA)
Wind speed	2000–2007	Daily	National Oceanic and Atmospheric Administration (NOAA)
Fine mode fraction (FMF)	2000–2009	Monthly	The Moderate Resolution Imaging Spectroradiometer (MODIS) Retrieval/Terra
Aerosol optical depth (AOD_{sat})	2000–2009	Monthly	
Ångström exponent	2000–2009	Monthly	
Aerosol optical thickness ($AOD_{aeronet}$)	2000–2007	Monthly	Aerosol Robotic Network (AERONET)

2.2. Data Sets. This work covers ten years period from 2000 to 2009 using ground and satellite observations to analyze the existing patterns and origin of pollutants over Seoul in relation to the prevailing meteorology [34]. Ground observation data, including SO_2, NO_2, PM_{10}, and CO, were measured at 44-air pollution monitoring stations, managed by the National Institute of Environmental Research (Figure 1). Twenty-six stations of which are situated within the Seoul metropolitan area while the other eighteen stations surrounding Seoul were used for correcting edge effects of air pollutants over Seoul. At these stations, the data for the four air pollutants were continuously measured during the study period (Table 1).

Temperature and precipitation have been provided from 46 Automatic Weather Systems (AWS) located in the same boundary areas as air pollutants, by the Korea Meteorological Administration (KMA), between 2000 and 2007. Wind speed data have been supplied from the US NOAA only for three locations in west, south, and center during the same time frame as temperature and precipitation. Humidity data were available only at two locations, south from NOAA and center from AWS.

In this study, the satellite data used over Seoul covers the area with following coordinates: top 37.701364, bottom 37.428478, left 126.7642, and right 127.1835. AOD_{sat} is obtained at 550 nm from the Moderate Resolution Imaging Spectroradiometer (MODIS)/Terra and $AOD_{aeronet}$, at 440 nm, from Aerosol Robotic Network (AERONET) (Table 1). A high correlation coefficient of 0.84 is obtained between AOD_{sat} data retrieved from MODIS/Terra satellite and $AOD_{aeronet}$ collected at Seoul National University's AERONET site. The Ångström exponent and fine mode fraction (FMF) are used to distinguish aerosols' origin either natural such as dust events or anthropogenic [40]. The FMF is the fraction of the AOD_{sat} contributed to by fine aerosols and distinguishes how the aerosols were derived, that is, from dust or anthropogenic origins, by determining the size of the aerosols [51, 52].

3. Results

3.1. Air Pollutants and Meteorological Parameters. Meteorological parameters such as temperature, precipitation, and wind speed play a pivotal role in air pollutants dynamics and distribution in many different ways [53, 54]. Here, we have carried out correlation analysis between the four pollutants mentioned in Table 1 and the corresponding meteorological parameters, namely, temperature and precipitation, over the 8-year period, from 2000 to 2007, taking seasonality into consideration.

All pollutants showed a significant negative correlation with temperature and precipitation before removing seasonality than after removing it, yet with positive correlations among pollutants (Table 2), a result that was partially found by Park et al. [55]. To further investigate the pollutants dependencies on the seasonal component, we have carried another cross-correlation analysis between the aforementioned parameters during spring, summer, fall, and winter seasons (Table 3). The negative correlation coefficient between pollutants and precipitation during all seasons suggests that removal process takes place and affects deposition to a great extent. Yet, the fall season showed the least dependency between pollutants and the meteorological parameters, implying that the local meteorology does not affect pollution concentrations during the fall season. This is owed to the fact that during fall and winter the atmosphere is less stable as compared to summer resulting in more pollutants dispersion

Table 2: The correlation coefficients between temperature, precipitation, and air pollutants: (a) with seasonal effects included and (b) with seasonal effects removed.

(a)

	Pre	NO_2	CO	SO_2	PM_{10}
Temp.	0.59	−0.70	−0.72	−0.82	−0.43
Precip.		−0.66	−0.46	−0.55	−0.43
NO_2			0.61	0.72	0.69
CO				0.75	0.44
SO_2					0.44

(b)

	Pre	NO_2	CO	SO_2	PM_{10}
Temp.	−0.03	0.19	0.07	0.07	0.16
Precip.		−0.16	−0.13	−0.26	−0.06
NO_2			0.22	0.23	0.40
CO				0.39	0.39
SO_2					0.06

[56]. On the other hand, summer showed a high correlation between NO_2 and PM_{10} as compared to other pollutants during other seasons and this is owed to the high traffic experienced during summer.

The previous analysis was performed by averaging all the meteorological parameters and pollutant data over Seoul, yet we wanted to further investigate the dependence on different regions within Seoul. For that, we selected five sites at the four cardinal directions: east (127.1E, 37.5N), west (126.8E, 37.5N), south (126.9E, 37.5N), and north (127.04E, 37.6N), and at the center (126.9E, 37.6N). Each of these sites has different geological characteristics and processes, for instance, east, north, and south parts are surrounded by mountain and west is close to the ocean. In addition, vehicles and human activities vary; during the daytime the center is crowded as many companies and major offices are located within, yet, the other sites are mixed industrial and residential areas.

It is interesting that the annual mean values of CO and SO_2 at the central location decrease gradually to a minimum in 2003 and in 2002, respectively, where further investigation is needed.

Each pollutant showed different abundances over 10-year averaged concentration at each site. The CO concentration showed notable differences between pollutants. The highest CO concentration is shown at the center (0.89 ppm) then the order of south, west, east, and north (0.65, 0.62, 0.62, and 0.56 ppm, resp.). It could be due to heating from residences and vehicles, the main sources of CO emission. Generally the center and south experience heavy traffic more than the other sites and less residences and offices exist at the north. The PM_{10} concentration indicates the highest at the center as CO; however the south site takes place for the lowest. Unexpectedly the highest concentration of NO_2 and SO_2 occurred at the south site; it could be due to vehicles and industrial activities.

We added wind speed at the center, west, and south (no data for the east and north) sites. According to other studies concentration of the four pollutants, pollutants react sensitively with wind speed [57–59]. Moreover, wind plays an important role of air pollution dispersion [60]. However, the results show low correlation coefficient: only west site shows notable negative correlation with NO_2 (−0.45). The west site is the only one place that is not surrounded by mountains, while the other sites are crowed with skyscrapers. This indicates that geological and specific megacity's features are one of important elements to determine pollutants concentration.

3.2. Annual Mean Analysis of the Four Air Pollutant Concentrations. Various pollutants are emitted from many different sources in megacities including Seoul, where high temperature combustion processes, such as from power plants and automobiles, are the main emission sources of NO_2 that has been slightly decreasing over the last decade due to the combined effects of various control efforts. However, increasing trends were still observed in some locations with high concentrations of CO [61], derived from vehicles and forest fires as well as SO_2 emission from burning fossil fuel and some industrial activities [48]. On the other hand, PM_{10} emission is strongly connected to different types of natural (dust storms) and anthropogenic sources [48]. Therefore, the PM_{10} studies are associated with specific dust events [37, 62–64]: high occurrences during spring (about 87% of the annual frequency in Korea) (Figure 2).

The NO_2 annual mean in 2008 showed the highest peak (0.0363 ppm) and in 2002 and 2005 showed the lowest. The increasing population and number of cars are the reason for the variations of annual mean because vehicles contribute to more than 70% of NO_2 [48]. Figure 3 supports the variation of annual mean where the highest number of vehicles and population is experienced during 2008, low population and stable vehicles number during the 2003–2005 period, and lowest population but increasing vehicles in 2002.

The annual mean values showed that CO concentrations decreased much more than the other pollutants over the 10-year period. The maximum value (1.0 ppm) is shown in 2000 and all months observed the highest concentration with exception of March and April (the highest March and April value was in 2001). The lowest annual mean is observed during 2005 with 0.63 ppm. The CO monthly means showed high concentrations during winter and low concentrations during summer, is analogous to NO_2. These two pollutants show similar concentration tendencies in monthly mean, which is consistent with previous study [61].

SO_2 showed two peaks in 2000 (0.007 ppm) and 2007 (0.0065 ppm) whereas the lowest value is observed in 2002 (0.005 ppm). The annual concentration decreased rapidly from 2000 to 2002, which would suggest that the emission control policies are established since 1995. Nevertheless, it increased again after 2004.

In 2002, the highest PM_{10} annual value (70.41 $\mu g/m^3$) was recorded and thus could be attributed to dust events. Dust was transported from China to Korea on the 21st of March, with a concentration of 1,153 $\mu g/m^3$ (Figure 4) along with the prevailing wind direction from the west with speed 5.6 m/s. Kim [37] reported that dust events have significantly

Table 3: The correlation coefficients between temperature and precipitation and air pollutants by season (with seasonal effects).

	Spring					Summer					Fall				
	Pre	NO_2	CO	SO_2	PM_{10}	Pre	NO_2	CO	SO_2	PM_{10}	Pre	NO_2	CO	SO_2	PM_{10}
Temp.	0.55	−0.16	−0.38	−0.51	−0.22	0.16	−0.74	0.0	−0.32	−0.56	0.54	−0.77	−0.64	−0.79	−0.70
Precip.		−0.21	−0.47	−0.48	−0.16		−0.46	−0.14	−0.50	−0.43		−0.68	−0.46	−0.43	−0.53
NO_2			0.45	0.38	0.33			0.35	0.48	0.80			0.58	0.78	0.80
CO				0.65	0.49				0.30	0.52				0.69	0.74
SO_2					0.04					0.49					0.63

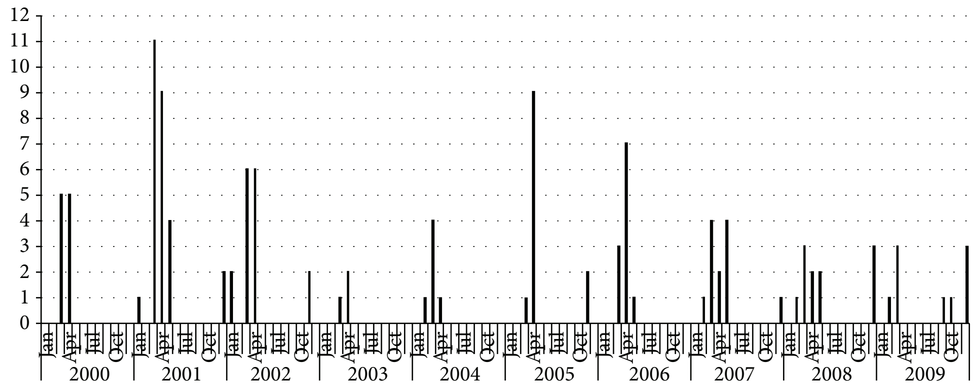

Figure 2: Dust event frequencies over the 10 years period.

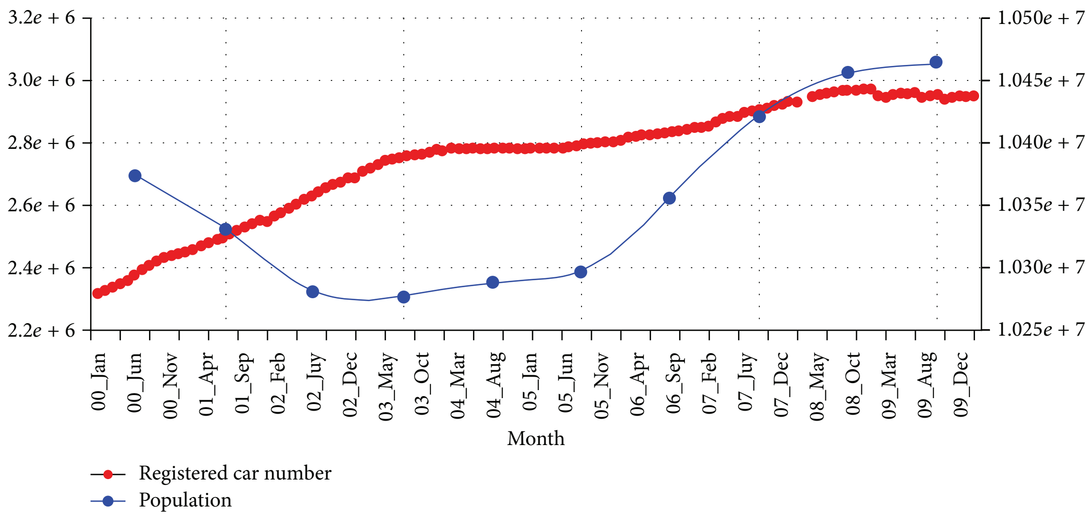

Figure 3: The number of registered car and population between 2000 and 2009.

increased during the 3 years from 2000 to 2002 and it has been increasing continuously since the 1980. The second highest peak was in 2007 with 62.74 $\mu g/m^3$, is much lower value than first peak even though dust events were more frequent in 2007 than in 2002.

3.3. Monthly Mean Analysis of the Four Air Pollutant Concentrations. The monthly NO_2 trends showed the same pattern as previous studies, that is, low concentrations during summer and high concentrations during winter [39, 54]. Figure 6(a) indicates that the NO_2 in first (2000) and sixth (2005) low peaks showed the lowest monthly mean values during the summer. On the other hand, NO_2 in ninth peak (2007) showed the highest monthly mean value. During the summer, the rapid decreasing concentration of NO_2 is due to atmospheric conditions such as amounts of humidity and precipitation [65, 66]: according to Haberer et al. 2006 [65]

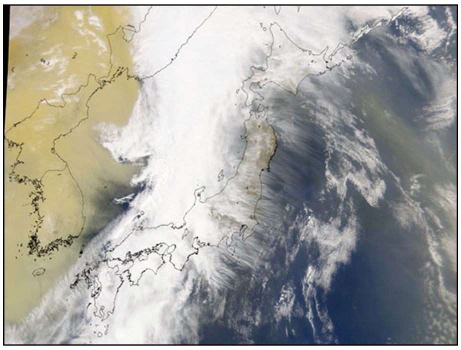

Figure 4: MODIS Terra image for 21 March, 2002, dust storm. Dust is lighter than the background in this true color MODIS-Terra image.

study, the minimum concentration of NO_2 investigated in July and NO_2 showed positive correlation with humidity. However, Korea experiences Monsoon every summer (this season takes most amount of annual precipitation); thus high humidity and huge amount of precipitation exist.

The CO monthly mean concentration showed the first peak during the winter of 2000 that was much higher than in the other years (Figure 6(b)). This could be due to the main sources of district heating and vehicular emission. According to the Ministry of Environment reports, the emission from vehicle takes a fist rank position and the consumption of fuel is the second reason of the entire CO emission [48]. The number of registered car has been increasing since 2000; however, the trend of oil consumption has been decreasing, exactly same pattern as the CO annual mean, except for the year 2000 (Figure 5). It indicates that CO concentration is significantly dependent on human activities.

The monthly pattern of the SO_2 concentration mostly showed the same behavior as that of CO and NO_2, with the highest concentrations during winter and the lowest during the summer (Figure 6(c)). According to Lalas et al. [67], in Greece, a low concentration was observed in late summer and early fall because of rain combined with a deep mixed layer. We mentioned above that the monsoon influences Korea during the summer; thus precipitation could explain that the low concentration was observed during summer and could be related to amount of rain and it reacts with precipitation dissimilar to other pollutants.

Figures 4 and 6(d) show that the 2002 dust storm was recorded as an intense event much stronger than other dust events in other years [68, 69]. The monthly mean of PM_{10} during spring season always takes high values during a year; during the winter it is of higher values than summer and fall seasons. However, the lowest concentrations always occurred during summer season due to the wash-out effect of summer rainfall: the lowest values were recorded in August in 2008, at 31.96 $\mu g/m^3$. Dust storms were definitely one of the main sources of high PM_{10} concentration [70, 71] during spring season. Sabbah and Hasan [72] found that the increase in wind speed is related to the increase of the concentration of dust particles during spring over the Solar Village (Riyadh, Saudi Arabia). Generally, during the spring the wind blows strongly from west to east over Korea.

3.4. Analysis of Pollutant Sources. In Sections 3.2 and 3.3, we show temporal characteristic of each pollutant with some possible reasons for 10 years period in Seoul metropolitan area. However, it has limitation of recognizing their origin; therefore we used satellite data (AOD_{sat}, Ångström exponent, and FMF) to distinguish their origins.

The satellite results indicate that AOD_{sat} increased from March to May, as expected due to the dust storms during the spring. All years during this time show AOD increasing with both low and high Ångström exponent and FMF representing a possible mixing scenario. During March 2002, a strong dust storm occurred (Figure 4), where the Ångström exponent, AOD_{sat}, and FMF values were 0.9, 0.45, and 0.1, respectively (Figure 7). These values suggest that this dusty event contains aerosols derived not only from natural sources but also from anthropogenic ones.

Unlikely, all Ångström exponent values were higher than 1 with decreasing AOD_{sat} values, while the FMF showed highest values (Figure 7) during the summer. For better understanding of distribution and origin of air pollution, we categorized the condition of atmospheric using AOD_{sat} and Ångström exponent values as same as "clean" with AOD $\leq$ 0.06, "dust" with AOD > 0.06 and Ångström exponent < 0.25, and "pollution" with Ångström exponent > 1.0, and other cases are classified as "mixed" [21]. The results show that the conditions of atmospheric over Seoul only have the cases of "mixed" and "pollution." In addition, most of the pollution occurs from May to October and mixed condition exists from November to April.

3.5. Volume Size Distribution Variability. Here we investigate the variation of the volume size distributions (VSD) as a function of the particle geometric mean radius for Anmyeon located 180 km from Seoul during the period 2000 to 2007 except for 2003 over west-ward direction as well as over Seoul for 4 years from 2000 till 2003 (Figure 8). The VSD was inverted from the AERONT observations at Anmyeon and Seoul, it helps understand local aerosol properties and variation [73, 74]. Figure 8(b) displays the variation of the VSD ($dV/d\ln R$) with a high magnitude of coarse mode peaking during the years 2005, 2006, and 2007 as compared to the previous years. The year 2002 has the highest concentration of coarse particles at Anmyeon due to the dust storm of 21 March, 2002. It peaks at ~3 μm geometric mean radius with partial contribution for the fine particles fraction.

We got a good correlation ($r = 0.60$) between the monthly values of AODaeronet collected at Seoul National University's AERONET site and the corresponding PM10 values during 2002, when a major dust event occurred, observed close to Seoul National University (Latitude: 37.45, Longitude: 126.9), and a correlation coefficient of −0.45 between Angstrom exponent and PM10. Sabbah et al. 2012 [75] found a good inverse correlation ($r = -0.62$) between the Ångström exponent and AOD_{sat} using Terra/MODIS satellite collections. They interpreted that as evidence of high

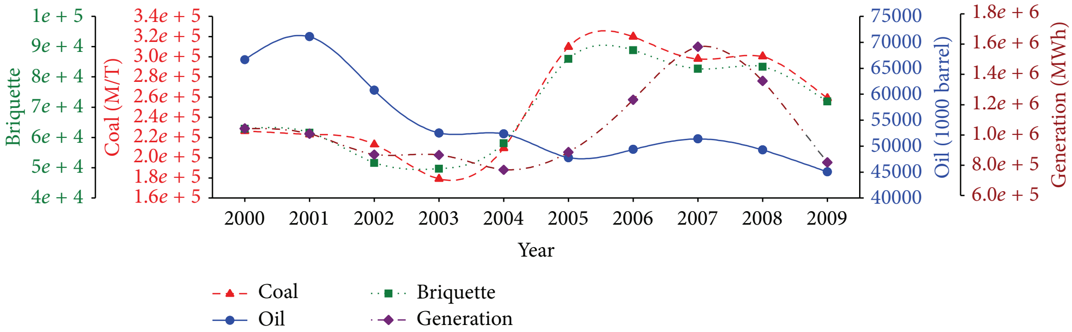

Figure 5: Annual means of coal and oil consumption and yield of briquette and generated.

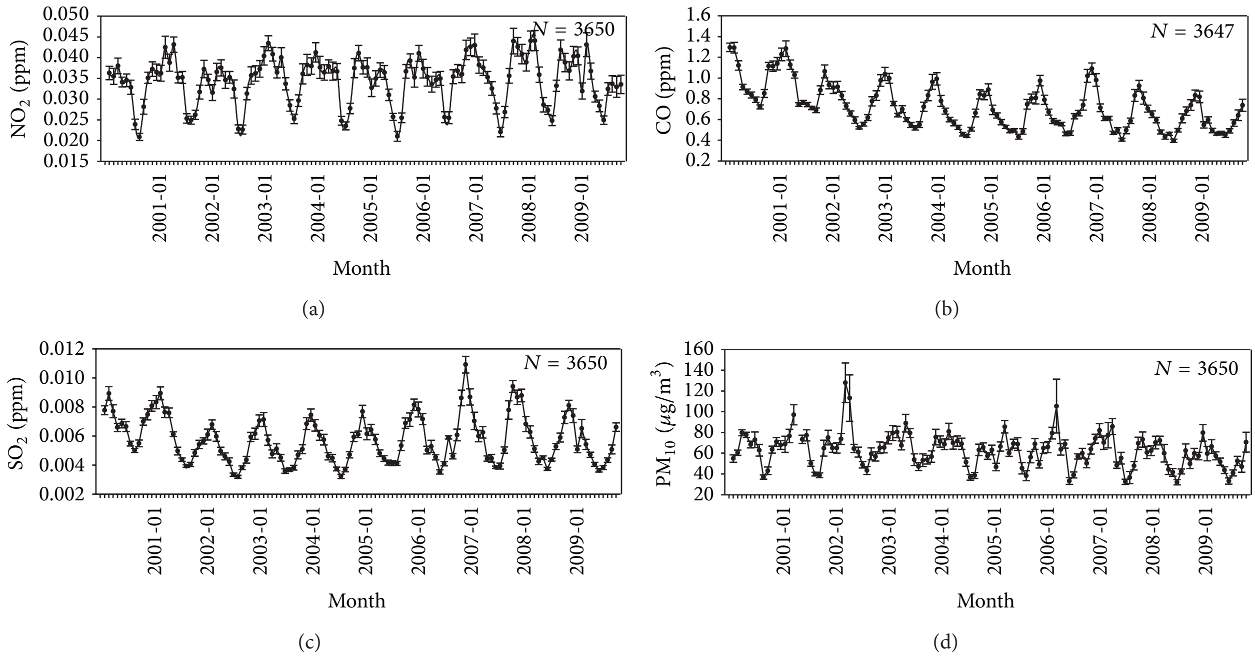

Figure 6: Monthly mean variations of (a) NO_2, (b) CO, (c) SO_2, and (d) PM_{10}between 2000 and 2009.

dust concentration. It supports that the dust storm in 2002 was high dust concentration. The year 2004 over Anmyeon exhibits the highest concentration of fine particles where the VSD peaks at 0.1 μm. Figure 8(d) illustrates the variation of the volume size distributions over Seoul for dust storm occurring on 21 March, 2002. We see a substantial increase in the concentration of the coarse particles at 2.9 μm that dominates the aerosol loadings on March 21 as evident from the major dust event over Seoul (Figure 4).

4. Discussion and Conclusions

In this study, major air pollutants over Seoul, including NO_2, CO, SO_2, and PM_{10}, were examined, revealing specific tendencies and characteristics, which can be attributed to natural and anthropogenic sources, as well as changing meteorological factors. The significant relationships between air pollution and temperature/precipitation exist due to the seasonality particularly; the strongest impacts were observed during the fall.

The annual mean results showed peak in different year related to different anthropogenic sources. This is evident from the increase in the cumulative number of registered car per month, rapid increase in population, high coal consumption, and the briquette yield. The dust storms, during the spring and winter, indicate mixed or pollution conditions. It means that even though dust storm contains not only natural aerosols but also aerosols from anthropogenic sources [15, 20, 76, 77]. Therefore, the reason for the peaks and minimum values are due to both natural and anthropogenic sources playing a role in the same year. The atmospheric condition during the summer is clearly identified as pollution, although all four pollutants show the lowest concentration. On the other hand, during the fall (highly affected by seasonality), it showed the mixed condition.

Satellite data analysis distinguished the pollutants origins and VSD. It indicates that the year 2007 contains small

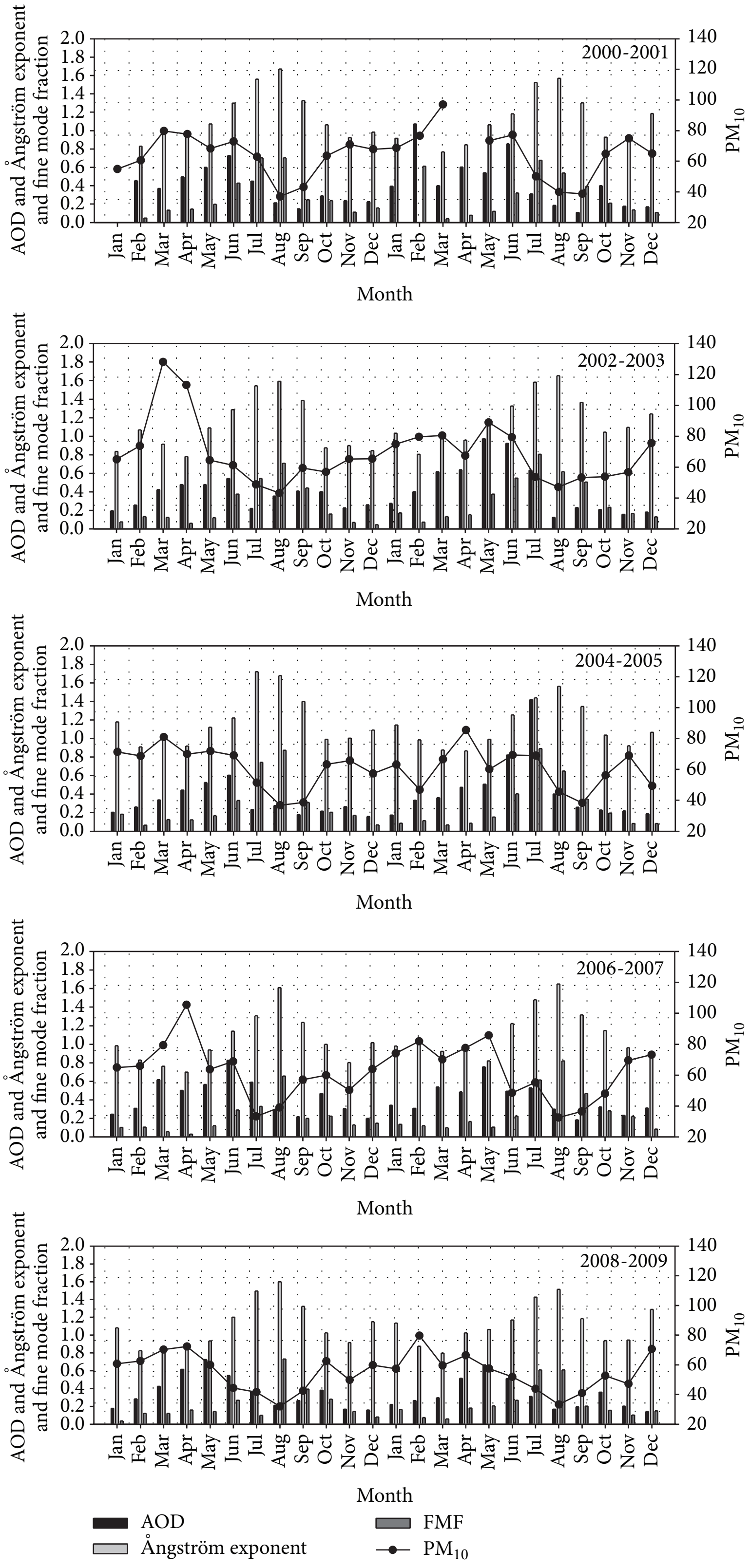

FIGURE 7: Satellite data (AOD_{sat}, Ångström exponent, and FMF) with PM_{10} (2000 to 2009).

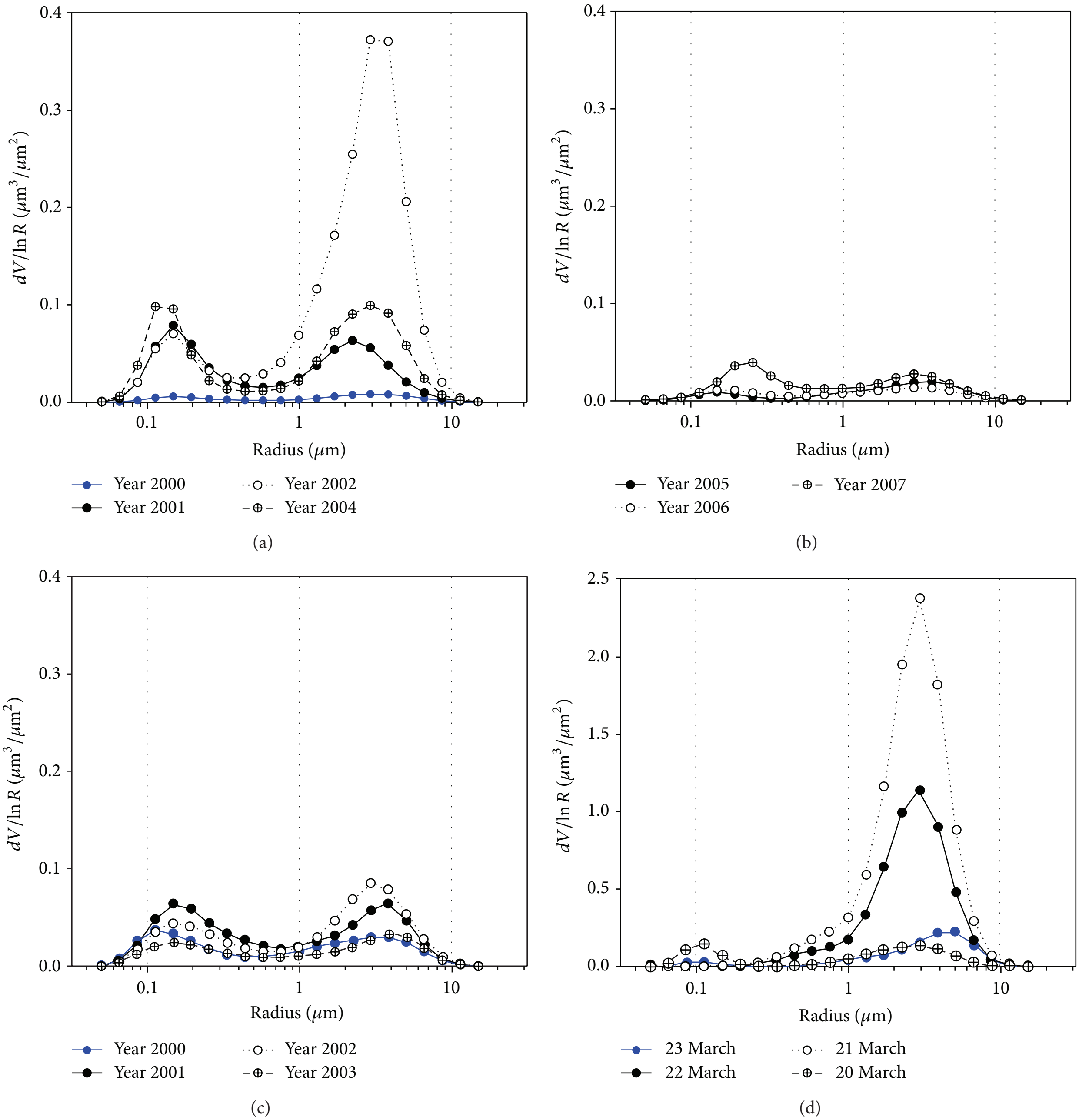

FIGURE 8: Variation of aerosol volume size distribution: (a) annual variation over Anmyeon for N = 63, 2000, N = 177, 2001, N = 42, 2002, and N = 209, 2004, (b) over Anmyeon for N = 117, 2005, N = 174, 2006, and N = 189, 2007, (c) over Seoul from for N = 15, 2000, N = 8, 2001, N = 106, 2002, and N = 14, 2003, and (d) daily variation over Seoul before, during, and after the dust storm of 21 March 2002. N is the number of days used.

size particle more than the other years: it implies that anthropogenic aerosol was predominant. In addition, anthropogenic aerosols increased in 2004 at Anmyeon rather than 2000, even though many policies have been established for air pollution: the radius of anthropogenic aerosols was found between 0.1 and 0.25 μm [78, 79]. The effective controls for air pollution in megacities, such as Seoul, must consider natural as well as anthropogenic sources when formulating laws to restrict pollutant emissions. Therefore, policy decisions need to be more elaborated to result in better air quality over Seoul.

Conflict of Interests

The authors declare that there is no conflict of interests regarding the publication of this paper.

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