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Ambient particulate matter (PM_{10}) concentrations in major urban areas of Korea during 1996–2010

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ABSTRACT

In this study, ambient particulate matter pollution was investigated using monthly PM_{10} concentration data collected from seven major cities in Korea from 1996 to 2010. The highest mean value for the whole study period is seen from the capital city, Seoul (63.2±17.9 $\mu g\ m^{-3}$), while the lowest is from Ulsan (46.7±14.8 $\mu g\ m^{-3}$). The concentrations of PM_{10} in all cities exhibited seasonal variations with the peak values occurring consistently in spring (March or April). The PM_{10} data in each city consistently exhibited strong correlations ($p<0.01$) with gaseous pollutants (SO_2, NO_2, and CO), except for O_3 ($p>0.05$). The analysis of long term trends of PM_{10} levels indicates a weak but consistent decline in concentrations in most cities with the relative average annual reductions of between 0.4 and 2.8% y^{-1}.

Keywords: *Regulated pollutants, PM_{10}, long term study, spatio–temporal variation, Korea*

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1. Introduction

Like many developing countries in Asia, Korea has witnessed a rapid increase in urbanization and industrialization over the past few decades (Lim, 2003). The development and expansion of urban areas has been the primary cause of the deterioration of air quality in urban areas (Lim et al., 2010). In order to improve air quality and protect public health, legislation has been established for air pollutants such as sulfur dioxide (SO_2), carbon monoxide (CO), nitrogen oxide (NO_2), ozone (O_3), PM_{10} (particulate matter with aerodynamic diameter <10 μm), and lead (Pb) (Neuberger et al., 2004; Le Tertre et al., 2005).

Many studies have demonstrated a close relationship between particulate matter (PM_{10}) pollution and deterioration in human health (Kappos et al., 2004; Wilson et al., 2005). The key properties of airborne particles are generally considered to be the size of aerosols and the associated capacity for penetration into the human respiratory system. This is supported by epidemiological evidence (Foster, 1999). As such, the concentration of PM_{10} has been monitored extensively in urban areas in many western countries (Bernardoni et al., 2011; Vicente et al., 2012) followed by many Asian countries (Wang et al., 2002; Lee et al., 2011).

In this study, the PM_{10} concentrations were investigated in the major urban areas of Korea using datasets collected between 1996 and 2010 (KMOE, 2010). These datasets were used to describe the general patterns of PM_{10} pollution in relation to both spatial (between cities) and temporal factors (e.g., seasonal and inter–annual variation). Based on these data, we analyzed the long–term trend of PM_{10} for each city using linear regression analysis. The level of PM_{10} pollution measured in major Korean cities has also been compared to other locations around the world.

2. Materials and Methods

2.1. Site characteristics of study area

To learn more about the environmental behavior of PM_{10} in major urban areas in Korea, its concentration data monitored continuously from seven major cities for the period of 1996–2010 were analyzed. Concentrations of PM_{10} and other key criteria pollutants were determined concurrently from urban air quality monitoring stations across seven major cities in Korea from 1996 to 2010. The geographical locations of those cities are described in Figure S1 (see the Supporting Material, SM) To facilitate comparison of the data across different cities the acronyms for each city were used (see the SM, Table S1): Seoul (SL), Busan (BS), Daegu (DG), Incheon (IC), Daejeon (DJ), Gwanju (GJ), and Ulsan (UL).

General information about the target cities has been described in our previous work (Nguyen and Kim, 2006). SL is the capital city with the largest metropolitan area in South Korea. BS is the second largest city with about 4 million people, serving as the country's main port for international cargo. (It belongs to the world’s fifth busiest port by cargo tonnage). DG is located in south–eastern part of Korea near Nakdong River, recording the Korea’s third most populous city. IC is a port city designated as the Korea’s first free economic zone in 2003. Since then a large number of local companies and global enterprises have increasingly invested in Incheon Free Economic Zone as the new investment destination. DJ had a population of over 1.5 million in 2010. The city is a hub of transportation due to the geographical location and is at the crossroads of major transport routes. UL is a highly industrialized city located on the south–eastern part of the Korea. UL is the industrial powerhouse with two enormous industrial complexes within the borders of the city, namely, the UL petrochemical complex and the UL Mipo industrial complex (Lee et al., 1999).

2.2. Data acquisition and management

As shown in Table S1 (see the SM), the number of individual urban air quality monitoring (UAQM) stations in all seven cities changed gradually each year from 51 (1996) to 95 stations (2010). According to KMOE protocol, each UAQM monitoring site is selected to be placed at less than 10 m height in the location of which monitoring is not disturbed by any physical barriers (buildings or trees). Each site should thus be placed to monitor the urban background air quality representative of the selected district. As such, our UAQM site is distinguished from other types of KMOE monitoring network such as Roadside monitoring site of which air quality is sensitively reflected by local traffic activity or National background monitoring site of which air quality is monitored from relatively remote locations (KMOE, 2010).

For the purpose of classification, all of the PM_{10} data collected from those individual stations were grouped into the seven major cities. After grouping, a total of up to 180 monthly PM_{10} data are available in each city for the whole study period (1996–2010). In the course of this study, PM_{10} values converted by the above procedure were examined further at various temporal scales (e.g., seasonal and inter–annual intervals) for each individual district. In addition, using monthly mean datasets for all different districts, correlation patterns were also analyzed and evaluated.

The PM_{10} data from each station were initially acquired at hourly intervals by the standard operating procedure of Korean Ministry of Environment (KMOE) based on the β–ray absorption method (Model FH62C14, Thermo Fisher Scientific, US). The PM_{10} data are measured at detection limit of ~5 μg m^{-3}. The hourly datasets collected from each monitoring station were ratified using the KMOE’s quality assurance procedure. The resulting data were then stored in the KMOE data management system and also converted into monthly averages (KMOE, 2010). Like the case of PM_{10}, all relevant parameters including criteria pollutants (NO_2, SO_2, O_3, etc) were measured and treated in the same manner described above. Details of data acquisition and treatment for those parameters have been reported elsewhere (e.g., Nguyen and Kim, 2006).

3. Results and Discussion

3.1. Spatial distribution pattern of PM_{10}

Statistical summary of the PM_{10} and relevant parameters measured at monthly intervals is presented (Table 1). To examine the spatial patterns of PM_{10}, the results were compared across different cities. The largest monthly PM value was found at SL (149 μg m^{-3}, March 2002), while the lowest was observed at UL (17 μg m^{-3}, August 1999). The mean PM_{10} values (μg m^{-3}) peaked in SL (63.2±17.9) followed by DG (62.2±17.3), BS (60.4±14.6), and IC (59.9±14.7), while the lowest values from UL (46.7±14.8) followed by DJ (49.8±14.0).

Table 1. *A statistical summary of PM_{10} and other relevant pollutant concentrations measured in seven major cities in Korea during the whole study period (using the monthly data measured between 1996–2010)*

	SL	BS	DG	IC	GJ	DJ	UL
				(A) PM_{10} data			
PM_{10} (μg m^{-3})	63.2±17.9[a] (64.0) 25.0–149.0[b] (179)	60.4±14.6 (58.0) 34.0–122.0 (180)	62.2±17.3 (61.0) 30.0–117.0 (177)	59.9±14.7 (57.5) 29.0–110.0 (180)	49.8±14.0 (51.0) 18.0–98.0 (178)	50.8±15.8 (51.0) 21–105.0 (175)	46.7±14.8 (45.0) 17.0–104.0 (166)
% decrease in PM_{10}	31.9	34.2	41.4	17.9	11.8	31.7	5.9
				(B) Other airborne pollutant data			
SO_2 (ppb)	6.6±3.1 (6.0) 3.0–19.0 (180)	9.4±5.6 (7.0) 3.0–31.0 (180)	8.9±5.9 (7.0) 3.0–36.0 (179)	8.0±2.8 (8.0) 4.0–22.0 (180)	5.1±2.4 (5.0) 2.0–14.0 (180)	7.1±9.3 (5.5) 2.0–120.0 (180)	11.6±5.0 (11.0) 4.0–30.0 (179)
NO_2 (ppb)	35.0±6.4 (35.0) 19.0–48.0 (179)	24.5±5.2 (24.0) 12.0–38.0 (180)	26.6±6.2 (26.0) 12.0–43.0 (177)	27.9±5.0 (28.0) 17.0–39.0 (180)	21.0±6.2 (21.0) 9.0–34.0 (177)	22.5±18.8 (21.0) 9.0–260.0 (180)	21.6±4.1 (22.0) 10.0–31.0 (177)
CO (ppb)	792.2±317.0 (700) 400–1 800 (180)	686.1±350.5 (600) 300–1 900 (180)	748.9±262.0 (700) 400–1 700 (174)	753.9±272.9 (700) 400–2 100 (180)	729.4±308.7 (700) 100–1 800 (180)	901.7±410.0 (800) 400–2 000 (180)	632.6±233.2 (600) 200–1 400 (175)
O_3 (ppb)	16.5±6.4 (16.0) 6.0–33.0 (180)	23.4±6.0 (22.0) 13.0–39.0 (180)	19.5±7.6 (18.0) 7.0–39.0 (179)	58.8±151.4 (18.0) 8.0–700 (180)	20.6±7.3 (20.0) 8.0–41.0 (180)	19.6±7.8 (18.0) 8.0–38.0 (179)	20.2±5.4 (19.5) 9.0–35.0 (180)

[a] *Mean±SD (Median), SD=Standard Deviation*

[b] *Range (Min–Max) (N), Min=Minimum, Max=Maximum, and N=Number of monthly data*

The enhanced PM_{10} values in SL and others (like DG, BS, and IC) may be attributed to the size of the city and the strengths of the related anthropogenic sources, as reflected by population size and traffic density (see the SM, Table S2). The observation of high PM_{10} levels in highly trafficked areas has been reported extensively. Based on the measurements at six locations of Kathmandu Valley, Nepal, Aryal et al. (2008) reported that the mean PM_{10} values varied considerably from 42 (rural area) to 230 μg m^{-3} (urban–roadside). Similarly, based on the study from Seoul, Korea, Bae et al. (2007) reported enhanced levels of PM_{10} in roadside (U–RS) over urban background (U–BG), despite similarity in relative diurnal patterns at each site. Likewise, Kim et al. (2010) also reported enhanced (24.5%) levels of PM_{10} at roadside relative to background site in Seoul which is explained by the direct effect and proximity of vehicular sources (Artinano et al., 2004). The relative ordering of mean PM_{10} values can be sorted on the order: UL<GJ<DJ<IC<BS<DG<SL (Table 1). Considering the patterns observed from many previous studies (Yang, 2002; Fuller and Green, 2006), information concerning the land use type can be used to explain at least partially the source characteristics of PM between different cities.

3.2. Temporal variability of PM_{10} between major cities

The PM_{10} data from each city were plotted at monthly intervals (Figure 1) across the whole study period. The PM data, when grouped by month and season (see the SM, Figure S2), indicated a consistent and systematic pattern with the observed seasonal values decreasing in the order: spring (March–May) followed by winter (December–February), fall (September–November), and summer months (June–August). Likewise, this trend has been commonly seen elsewhere, e.g., seven major cities of Taiwan during 2000–2008 (Fang and Chang, 2010). Comparison of the seasonal mean data shows the spring peak in SL (76 μg m^{-3}) with the summer minimum in DJ (40 μg m^{-3}).

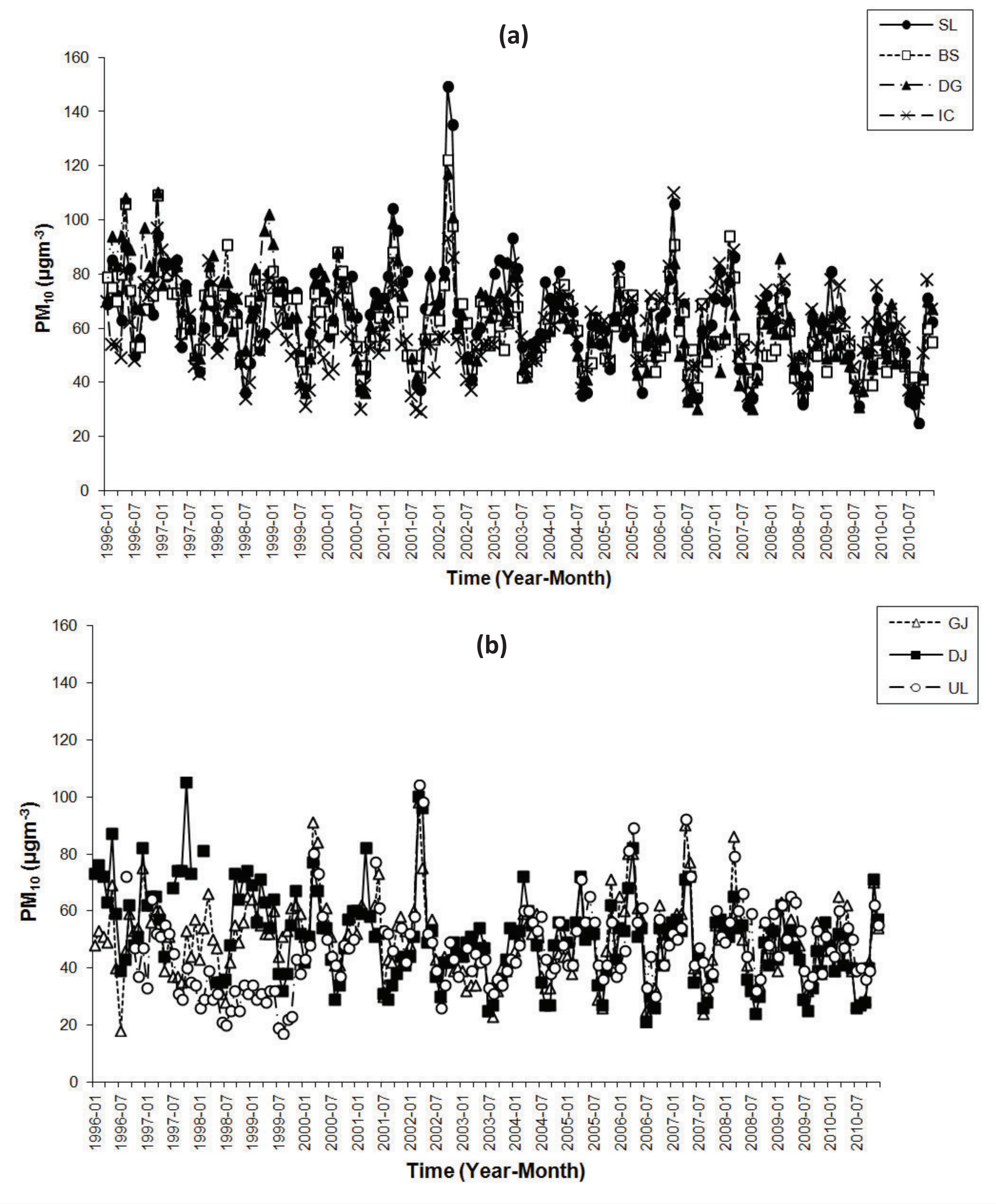

Figure 1. *Plot of monthly concentrations of PM_{10} (μg m^{-3}) monitored in seven major cities of Korea (1996–2010).* ***(a)*** *Four cities with higher PM_{10} concentrations;* ***(b)*** *Three cities with lower PM_{10} concentrations.*

The springtime peak of PM_{10} is coinciding with the Asian dust (AD) events that commonly occur during spring in East Asia (Kim and Kim, 2003; Lee et al., 2006). The total quantity of PM transported annually via AD events is estimated as 800 Tg (Zhang et al., 1997). The AD is known to transport suspended dust from deserts or loess in China and Mongolia to Korea by wind or turbulent flow (Lin, 2001) which can build–up the fine particle levels during spring (Kim et al., 2002). The number of springtime AD days accounted for 77% of total AD days during the study period (see the SM, Table S3). Recently, the AD days have increased in fall and winter. In contrast, the lowest PM_{10} concentrations in summer can be attributed to strong precipitation during monsoon (Song et al., 2012). The observed seasonality in PM_{10} is common for most East Asian countries (Massey et al., 2012).

To assess the variability of PM_{10} data during the study period, its relative amplitudes (RA in %) were computed using the monthly data: RA=[{(Max–Min)×100}/Mean]. Accordingly, the variabilities (RA) were found in the order of SL (220.9%), BS (156.7%), DG (155.0%), IC (144.3%), GJ (142.5%), DJ (149.6%), and UL (155.0%) to reflect the combined effect of city wide source processes and meteorological conditions prevailing in each area.

Information concerning the temporal trends of PM_{10} over the whole study period can be used to provide a general picture of the balancing mechanism between source/sink processes (Vicente et al., 2012). A better knowledge of balancing mechanism over a long–term period may allow assessment of the impact of policy–driven changes in pollutant levels, which can ultimately feedback into the formulation of more effective management strategies (Burt et al., 2008; Bahadur et al., 2011; Schichtel et al., 2011).

As seen in Figure 2, plots of long–term PM_{10} data indicate general reduction in its concentration levels. Although there are inter–annual variations in PM_{10} levels, we simply compared its reduction rate by comparing the concentration levels across the whole study period (e.g., between 1996 and 2010): 5.9% (UL) to 41.4% (DG) throughout the 15 year, e.g., SL from 72 (1996) to 49 μg m^{-3} (2010). The results indicate notable reduction in certain cities like DG (41.4%), while changes are insignificant in certain areas like UL (5.9%). Since 2000, the PM_{10} in SL continually increased to reach its maximum (76 μg m^{-3}) in 2002 and then decreased gradually to reach a minimum of 49 μg m^{-3} at 2010. The unusual rise in 2002 may reflect the severity of AD events in that year which took place 16 times. In April 2002, PM_{10} reached 3 311 μg m^{-3} in Hannam–dong in Seoul (NIER, 2004).

The long–term trend of PM_{10} was analyzed for each individual city by linear regression analysis using monthly PM_{10} concentration data of all period (N=166 to 180) (see the SM, Table S4). The results show a trend of gradual reduction (e.g., negative slope values) with a strong significance: SL (P<0.01), BS (P<0.01), DG (P<0.01), and DJ (P<0.01). However, in a few cities like IC (P=0.565) and GJ (P=0.129), the results are not systematic enough (0.1 and 1.3% variation, respectively). In case of UL, an upward trend was seen to reflect the effect of unusually low PM levels during 1998 and 1999 (Table 2). Although it is not evident to account for the cause of those patterns, the observed values during the two year period represent the least PM levels among all sites during the whole study period. In urban areas of Korea, emissions control efforts such as the enforcement of the Natural Gas Vehicle Supply (NGVS) program and emission control retrofits since June 2000 should have helped long–term reduction in PM_{10} levels (Kim and Shon, 2011). Decreasing PM_{10} levels in the urban environments have also been reported in many previous studies. For instance, Salvador et al. (2012) reported a decrease in PM_{10} concentrations from 48 (1999–2000) to 41 μg m^{-3} (2007–2008) in Madrid, Spain. This was also attributed to a reduction in the annual consumption of coal and in the number of gasoline vehicles. The use of particle filters for diesel engines would also have contributed to such trend (Salvador et al., 2011; Salvador et al., 2012).

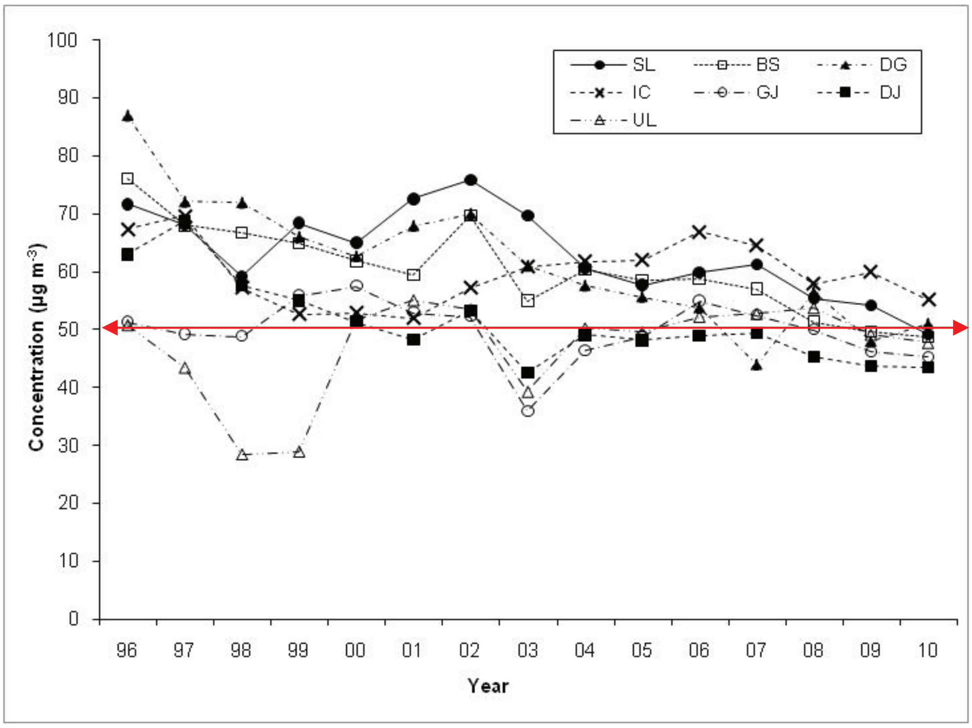

Figure 2. *Plot of the annual mean concentrations of PM_{10} (μg m^{-3}) from seven major cities of Korea over the entire study period (1996–2010); Red line: Annual average limit of PM_{10} as per the environmental policy regulation from Korean Ministry of the Environment (KMOE, 2009).*

Table 2. *Comparison of the PM_{10} concentration levels in this study and previous studies (in $\mu g\ m^{-3}$)*

City/Country	Ref [a]	Sampling Points	Annual Mean PM_{10} ($\mu g\ m^{-3}$)														
			1996	1997	1998	1999	2000	2001	2002	2003	2004	2005	2006	2007	2008	2009	2010
A. Asian Countries																	
A1. Urban Areas																	
Seoul, Korea	[1]	Citywide	72	68	59	68	65	73	76	70	61	58	60	61	55	54	49
Ulsan, Korea	[1]	Citywide	51	43	29	29	52	55	53	39	50	50	52	53	54	49	48
Dae Hung, Seoul, Korea	[2]	Single spot	52.1	87.3	90.0	70.4	64.9	73.0	78.4	73.1	39.4	43.5	52.9				
Banciao, Taipei, Taiwan	[4]	Single spot					57	49	45	48	61	58	51	52	49		
Taishi, Taiwan	[5]	Single spot	74.4	67.4	63.6	61.7											
Guanjin, Taiwan	[5]	Single spot	55.0	53.8	48.9	44.1											
URA, Kathmandu, Nepal	[6]	Single spot								149	139	122	124				
Agra, India	[7]	Single spot											160.2	148.2			
Kasba, Kolkata, India	[8]	Single spot								140							
Shanghai, China	[10]	Citywide	243	236	217	173	157	97	107	98	99	87					
Shanghai, China	[9]	Citywide											86	88	84	81	
Beijing, China	[10]	Citywide				180	162	164	165	141	148	142					
AECD, Dhaka, Bangladesh	[11]	Single spot						110	70.9	74.5							
Shin chon, Seoul, Korea	[2]	Single spot					76.4	70.0	87.5	78.8	72.2	65.1	59.3				
Farm gate, Dhaka, Bangladesh	[11]	Single spot						170	147	125							
BTUA1, Kathmandu, Nepal	[6]	Single spot								208	203	202	230				
BTUA2, Kathmandu, Nepal	[6]	Single spot								180	190	202	170				
A2. Rural and Suburban																	
Gosan, Korea	[3]	Single spot					29.6	32.3	26.3	22.9	32.0	38.0	27.6	20.7			
Erlin, Taiwan	[4]	Single spot					73	74	75	68	69	69	58	55	59		
Agra, India	[7]	Single spot											152.1	144.7			
Kathmandu, Nepal	[6]	Single spot								50	56	44	42				
SUA1, Kathmandu, Nepal	[6]	Single spot								80	78	65	62				
SUA2, Kathmandu, Nepal	[6]	Single spot								131	101	96	75				
A3. Industrial																	
Nantz, Taiwan	[5]	Single spot	89.5	77.9	65.9	78.9	79.7										
Linyuan, Taiwan	[5]	Single spot	110.8	83.8	75.5	76.1	94.5										

***Table 2.** Continued*

City/Country	Ref [a]	Sampling Points	Annual Mean PM_{10} ($\mu g\ m^{-3}$)														
			1996	1997	1998	1999	2000	2001	2002	2003	2004	2005	2006	2007	2008	2009	2010
B. European Countries																	
B1. Urban																	
CS, Palermo, Sicily, Italy	[12]	Single spot		46.3	43.8	43.9	46.5	43.8	43.1	40.3	39.3	35.5	40.4	37.7	36.8		
Rotterdam, Netherlands	[13]	Citywide	43.0	36.1	35.3	33.2	30.1	29.8	32.2	33.0	28.1	27.9	30.3	27.2	25.0		
Uskudar, Istanbul, Turkey	[15]											33.0	41.0	69.0	47.0	38.0	
Olivais, Lisbo, Portugal	[16]	Single spot						33.1	28.8	29.2	31.9	30.3	30.3	28.0			
Loures, Lisbo, Portugal	[16]	Single spot							35.7	36.3	32.4	32.8	31.1	28.2			
Inner London, UK	[17]	Citywide		8.5	5.5	6.5	5.5	6.5	6.3	7.7							
Bern, Switzerland	[18]	Single spot			40.3	37.9	33.0	32.5									
Lugano, Switzerland	[18]	Single spot			35.7	30.9	33.8	31.8									
Madrid basin, Spain	[19]	Citywide				48.0									41.0		
DB, Palermo, Sicily, Italy	[12]	Single spot							49.5	46.3	43.3	43.3	58.6	45.6	42.2		
Rotterdam, Netherlands	[13]	Citywide	45.0	40.5	34.4	32.5	31.2	31.3	32.5	35.5	32.6	31.5	32.6	30.7	25.6		
Besiktas, Istanbul, Turkey	[15]	Single spot										58.0	74.0	66.0	54.0	49.0	
Espinho, Porto, Portugal	[16]	Single spot							51.4	54.0	53.1	47.3	42.6	43.5			
Aveiro, Portugal	[16]	Single spot								44.1	38.1	38.3	33.8	40.9			
Central London, UK	[17]	Citywide			15.7	18.4	16.4	16.3	17.0	17.5							
B2. Rural and Suburban																	
Ìlhavo, Aveiro, Portugal	[16]	Single spot								34.9		28.0	28.1	28.1			
Chaumont, Switzerland	[18]	Single spot			10.6	12.1	10.2	11.0									
Payerne, Switzerland	[18]	Single spot			23.2	20.6	19.8	19.3									
Payerne, Switzerland	[18]	Single spot			23.2	20.6	19.8	19.3									
Estarreja, Portugal	[16]	Single spot							31.5	42.0	42.0	40.6	35.4	38.0			
Leça do Balio, Portugal	[16]	Single spot						54.4	43.9	45.9	36.8	36.1		38.5			
Athens area, Greece	[14]									44.7	43.8	40.6	33.4	29.3	30.6		
Outer London, UK	[17]	Citywide		5.9	3.5	4.4	3.9	4.9	4.8	5.9							
Dübendorf, Switzerland	[18]	Single spot			26.7	23.6	20.8	20.6									
Basel, Switzerland	[18]	Single spot			24.1	23.1	20.5	22.2									
BF, Palermo, Sicily, Italy	[12]	Single spot		27.2	24.5	23.2	24.4	25.5	28.1	29.2	28.1	22.3	23.2	23.2	22.2		
B3. Industrial																	
Kartal, Istanbul, Turkey	[15]	Single spot										72.0	102.0	82.0	70.0	72.0	
Yenibosna, Istanbul, Turkey	[15]	Single spot										47.0	53.0	73.0	72.0	62.0	

[a] *Source of reference information: [1] This study; [2] Kim et al., 2010; [3] Kim and Shon, 2011; [4] Fang and Chang, 2010; [5] Yang, 2002; [6] Aryal et al., 2008; [7] Kulshrestha et al., 2009; [8] Karar and Gupta, 2007; [9] Li et al., 2012; [10] Chan and Yao, 2008; [11] Begum et al., 2006; [12] Dongarra et al., 2010; [13] Keuken et al., 2011; [14] Pateraki et al., 2010; [15] Unal et al., 2011; [16] Borrego et al., 2010; [17] Fuller and Green 2006; [18] Gehrig and Buchmann, 2003; and [19] Salvador et al. (2011, 2012).*

3.3. Factors affecting the distribution of PM_{10}

A correlation analysis was conducted between: (1) PM levels between different cities and (2) PM_{10} vs. other parameters. The results in Table S5a (see the SM) indicate a strong correlation of PM_{10} levels between different cities (e.g., $P<0.01$). Consequently, the combined effects of some important variables affecting the PM distributions (e.g., consistency in their seasonalities and communalities in source processes (like traffic activities and AD events)) are likely to share common trends across different locations. In Korea, the contribution of primary (direct emission) and secondary sources (e.g., photochemical production) to the total mass of PM_{10} is generally estimated to comprise of 70% and 30%, respectively (Song and Shon, 2008; Kim and Shon, 2011). Hence, it is reasonable to infer that the role of direct emissions should be more important to explain the trends in PM_{10} between the cities.

The relationship between PM_{10} and anthropogenic emission (energy production, non–industry, manufacturing, mobile, non–mobile, and waste treatment) is examined in Figure 3. The anthropogenic PM_{10} emission in major cities was taken from NIER (http://airemiss.nier.go.kr/main.jsp). The major sources of PM_{10} emission in the cities were mobile and non–mobile sources, except for UL. The PM emissions from combustion (manufacturing and production process) were the dominant contributors in UL. If the correlation analysis is made additionally using the two variables shown in Figure 3 (i.e., annual mean values of PM emission and concentration), their correlation patterns was greatly distinguished by such variable as the size of city (or population). For instance, the stronger correlations were evident in SL (r of 0.77) followed by DG (0.51), BS (0.50), and IC (0.47), whereas it was insignificant in DJ (0.24), GJ (–0.12), and UL (0.04). The correlations between ambient PM_{10} concentrations and emissions of other pollutants (CO, SO_2, NO_x, and VOCs) were also examined. Although PM_{10} was generally correlated with CO [e.g., r(SL) and r(BS) >0.6) and NO_X [r(SL) and r(IC) >0.6], it was not with SO_2 (except for DG, r of 0.85) and VOCs (except for IC, r of 0.63). The strong correlation of PM emission with CO and NO_X in highly populated cities (SL, BS, are IC) is likely to be related to the traffic emission sources.

In Table S5b (see the SM), the correlations between PM_{10} and other parameters are compiled for each city. PM_{10} generally showed significant correlations ($P<0.01$) with most pollutants other than O_3 ($P>0.05$). It thus suggests that all three species (PM_{10}, CO, and NO_X) share similar source profiles, such as traffic emissions (Smith et al., 2001). Moreover, a strong correlation with SO_2 also suggests other sources like industrial activities (Sharma and Tripathi, 2009). Shon et al. (2012) suggested a significant role for the gas–phase oxidation of SO_2 (H_2SO_4) in SO_4^{2-} formation in light of the SO_4^{2-}/SO_2 mass ratio in SL. They identified the relative dominance of secondary inorganic ions such as NO_3^-, SO_4^{2-}, and NH_4^+ in fine particles. As such, the secondary formation of inorganic components in PM_{10} can be important enough to discriminate the PM_{10} distribution patterns between different cities (Ansari and Pandis, 1998).

3.4. Comparison of PM_{10} levels between different studies

The PM_{10} values in our study were compared with those made previously in Korea and abroad (Table 2). If our annual mean PM_{10} values are compared with the guideline of the KMOE (2010), the values at IC exceeded the permissible limit (i.e., ≤50 μg m^{-3}) consistently through the study period. Similarly, PM_{10} values above the guideline were also seen at DG, BS and SL. The results of different studies were compared after being grouped into different land use types (urban, rural and suburban, and industrial) (Table 2). For the sake of simplicity, the results of SL and UL were taken as representatives of our study for comparison.

Although high PM_{10} level was recorded in Shanghai in the late 90s, it dropped notably by 2000 and then continued to show a steady reduction until recently reaching 81 μg m^{-3} (Chan and Yao, 2008; Li et al., 2012) (see the SM, Figure S3a). The annual mean of PM_{10} in SL between 1999 to 2005 (67.3 μg m^{-3}) was found approximately 1.7 and 2.3 times lower than in Shanghai (116.9 μg m^{-3}, 1999–2005) and Beijing (157.4 μg m^{-3}, 1999–2005), respectively (Chan and Yao, 2008). This type of comparison has been extended further to the values in European countries under well advanced emission legislation (Italy and Netherlands) for the same period (1997–2008) (see the SM, Figure S3b). It was noticed that in Palermo (Italy) and Rotterdam (Netherlands), it decreased rapidly in the early years (1996 to 2001) and then began to show more fluctuation without clear trends (2003 to 2008). Such patterns are apparent in recent years in many European cities that trend to maintain low annual mean PM_{10} values, such as in London, UK, Lisbon, Portugal, Athens, Greece, and Dubendorf, Switzerland (e.g., mean of 15.0 to 30 μg m^{-3}, Table 2).

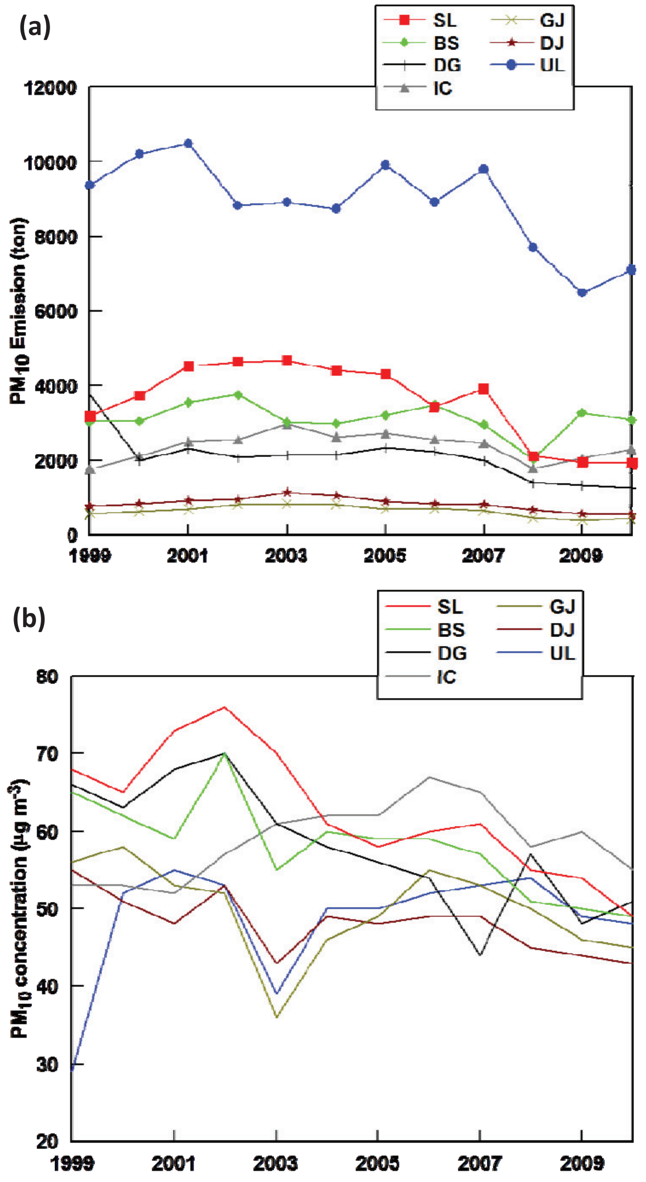

Figure 3. (a) *Plot of the annual anthropogenic PM_{10} emission (tonnes) and* ***(b)*** *annual PM_{10} concentration (μg m^{-3}) from seven major cities of Korea over the entire study period (1996–2010).*

PM_{10} levels in SL (55 μg m^{-3}, 2008) are approximately 2.2–2.5 times greater than their European counterparts such as the Netherlands (25 μg m^{-3}, 2008) and Italy (22.2 μg m^{-3}, 2008). Moreover, PM_{10} concentrations in most of the European cities would fall well below the permissible limit of KMOE (<50 µg m^{-3}) with some exceptions, e.g., Turkey and Portugal (Table 2). However, most of these cities would still exceed the stricter World Health Organization guideline of 20 µg m^{-3} (WHO, 2006).

4. Conclusions

PM_{10} concentrations were investigated using the monthly datasets measured from air quality monitoring stations in the seven major cities of Korea during 1996 to 2010. Its spatial distribution patterns showed the maximum value of 63.2 µg m^{-3} in the capital city, SL and the lowest value (46.7 µg m^{-3}) at the coastal city, UL. The analysis of seasonal patterns indicates the springtime peak to reflect the combined effects of source processes like Asian Dust and meteorological conditions.

The analysis of long–term trend indicates that it decreased consistently in six out of all seven cities during the years 1996–2010. Such consistency can be observed across the Korean peninsula, reflecting the effects of the emissions control efforts.

If the status of PM_{10} pollution in major cities of Korea is compared with the guidance level of KMOE (i.e., ≤50 µg m^{-3}), the results generally showed exceedance in most cities in earlier years. However, the most recent values (e.g., in 2010) in most cities complied with this limit value (except DG and IC). Comparison of the PM_{10} levels of the present study with those measured from the other cities in Asian and European countries confirms that there are apparent spatial gradients in PM_{10} concentration levels between continents and between areas of different land use types. However, PM_{10} concentrations in most areas generally exhibit consistent decreases during the last 15 years.

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Supporting Material Available

The number of individual air quality sampling stations in seven major cities of Korea during the whole study period (1996 to 2010) (Table S1), The statistics of the major cities in Korea: Population density (in thousand persons) and the number of vehicles registration as of 2010 (Table S2), The number of days of Asian dust event in each month of the year during the whole study period in Seoul (Table S3), Result of linear regression analysis using the monthly PM_{10} values measured during the entire study period (1996–2010) in each of all seven cities (Table S4), Results of correlation analysis using the monthly PM_{10} data (Table S5), A map showing the locations of all seven cities in Korea investigated in this study (Figure S1), Temporal trend of PM_{10} levels (µg m^{-3}) across the major cities of Korea: (a) monthly and (b) seasonal scales (Figure S2), Comparison of long–term PM_{10} concentration measured in urban and suburban areas of Asia and Europe for the same period (Figure S3). This information is available free of charge via the internet at: http://www.atmospolres.com.

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