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Long-Term Variations of Airborne Cadmium (Cd) Concentrations in Major Urban Areas of Korea between 1991 and 2010

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ABSTRACT

The long-term (1991–2010) monitoring data for cadmium (Cd) collected from seven major cities in Korea were evaluated to assess spatial-temporal trends in urban areas and help improve air quality control. The results show that the level of Cd pollution in the different cities, as distinguished by industrial activity, was the highest in Ulsan (9.21 ± 6.29 ng/m^3) and Incheon (6.25 ± 6.05 ng/m^3). During the study period, Cd concentrations showed significant reductions in all monitoring cities, especially in the more industrialized ones (e.g., a nearly six-fold reduction in Ulsan over 13 years), while in other areas this reduction was more moderate (about 1.7-fold). In addition, Cd concentrations generally peaked in the winter, with the exception of the more industrialized areas. Although Cd levels in Ulsan and Incheon exceeded the general guidelines of 5 ng/m^3 (e.g., those of the US EPA) until recently (e.g., 2004), its values in most urban areas in Korea have been continuously decreasing over the 20-year period examined in this work. Therefore, greater efforts are necessary to more effectively reduce Cd released from the prominent source activities in the urban environments.

Keywords: Cadmium (Cd); Long-term; Spatiotemporal; Industrial; Urban.

INTRODUCTION

Like most airborne metallic pollutants, cadmium (Cd) is released from both natural and anthropogenic sources and present in a particle bound form in ambient air. The dominant fraction of airborne Cd generally comes from anthropogenic processes such as non-ferrous metal production, the mining and refining of zinc (and lead) ores (Bernard, 2008), the combustion of fossil fuels, and the operation of waste incinerators and power plants (USEPA, 2000; ATSDR, 2008; Avino *et al.*, 2008; UNEP, 2010). The use of Cd can be expanded further to a stabilizer in the production of batteries, pigments, polymers, and plating (USEPA, 2000; ATSDR, 2008; Avino *et al.*, 2008; UNEP, 2010). In our everyday activities, the intake of Cd can also occur through such activities as direct or indirect exposure to tobacco smoke (ATSDR, 2008). In contrast, terrestrial dust, wild fires, and volcanic activity are classified as the main sources of natural Cd (Avino *et al.*, 2008; Hieu and Lee, 2010).

Cadmium (Cd) has drawn most environmental concern with respect to significant ecological risk due to high toxicity and mobility (Bi e*t al.*, 2006; Kim *et al.*, 2003, 2009; Ma *et al.*, 2012; Ray *et al.*, 2012). The US EPA (2011) has classified airborne cadmium as a probable human carcinogen (Group B1) listed substance (with limited evidence of carcinogenicity). Cadmium, if exposed chronically, may cause proteinuria and/or damage lungs due to bronchial and pulmonary irritation (USEPA, 2000; Glorennec *et al.*, 2005; UNEP, 2010; USEPA, 2011). Furthermore, it can be transferred to food chain due to its high potential for accumulation in various parts of the ecosystem (soil, sediment, water, and plants) (Bernard, 2008; Aas *et al.*, 2009). Because of great efforts with the support of advanced control technology, a constant reduction in the concentration levels of many toxic metal pollutants is observed in most of world including North America and Western Europe (e.g., Harmens *et al.*, 2007).

To assess the long-term variability of airborne Cd levels in urban areas, we analyzed the airborne Cd concentration data acquired from the seven major cities of Korea over a 20-year period (1991–2010) by the Korean Ministry of Environment (KMOE). As the long-term monitoring of Cd was made at each of the seven major cities in Korea, these Cd data can be examined to assess its spatiotemporal variability in a number of respects. The results of our analysis will thus help us learn more about the fundamental characteristics of Cd in the urban environment and develop strategies to control its pollution. In our previous study, the data sets of Cd collected during the earlier period of this study (1991–2004) had already been evaluated (Kim, 2007). Hence, the results of this study will offer us an opportunity to assess the temporal trends of Cd over an extended period up to 2010.

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MATERIALS AND METHODS

In this study, we analyzed the Cd data measured from 7 major cities (Seoul, Busan, Daegu, Incheon, Gwangju, Daejeon, and Ulsan) in Korea between 1991 and 2010 (Fig. 1). These seven major cities are well-known for their high population densities, large urban residential areas, and major industrial (and transportation) activities. The overview of the size, population, temperature, and humidity in the major monitoring areas are shown in Table 1.

To assess the extent of metal pollution in the urban atmosphere, concentrations of hazardous metal species have been measured routinely from major urban areas of Korea with large populations (e.g., above a half million) or strong industrial activities. As part of such efforts, Cd concentration data measured from seven major cities in Korea between 1991–2010 were evaluated to account for the basic status of Cd pollution over an extended period. Seoul had the largest number of monitoring stations totaling 13 in 1998 and was followed by Daejeon (7), Busan (5), Daegu (5), Gwangju (4), Ulsan (4), and Incheon (3). At each individual station, the collection and analysis of particle samples were made by following the basic analytical procedures recommended by the air quality measurement method of the KMOE. According to this method, the amounts of all metals are determined from total suspended particle (TSP) samples collected by high-volume samplers with the aid of flame atomic absorption spectrometry (FAAS) (e.g., Kim *et al.*, 2004; Mutlu *et al.*, 2012). The Cd concentrations of the extracted solutions were determined in reference to a standard solution of trace metals certified by the US National Institute of Science and Technology (US NIST).

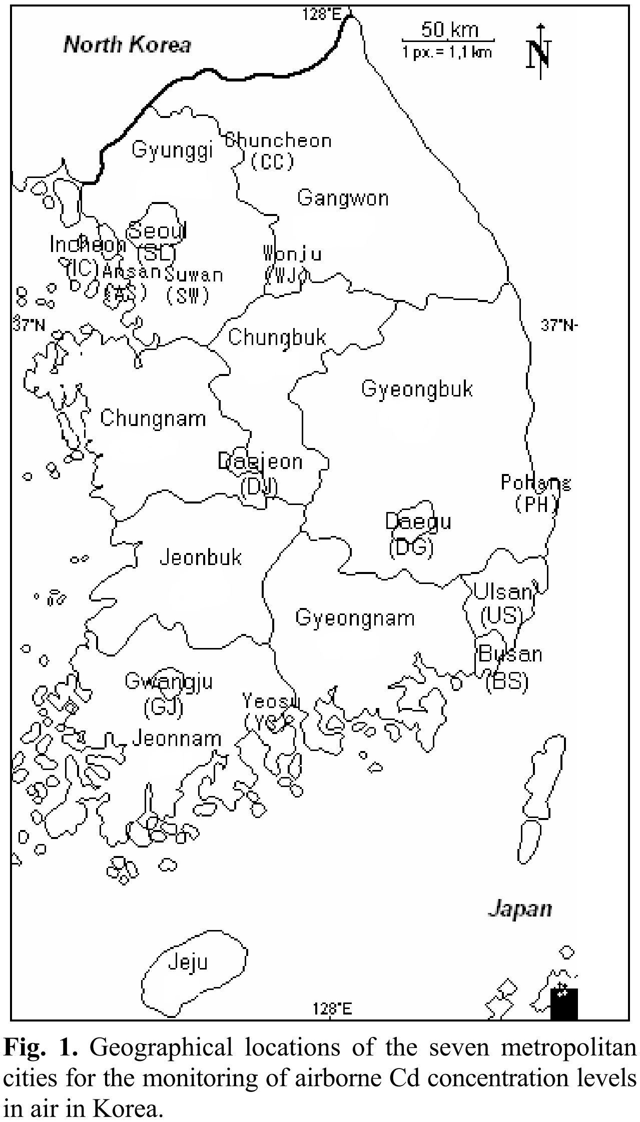


Fig. 1. Geographical locations of the seven metropolitan cities for the monitoring of airborne Cd concentration levels in air in Korea.

These original metal concentration data are then examined further by the quality assurance (QA) procedures of KMOE and stored in its data management network system. As the part of QA procedure in the analysis of Cd (and other metals), the reliability of the experimental method was examined at least on a once-per-year basis. The environmental behavior of airborne Cd measured for the entire 20-year study period (1991–2010) was evaluated after grouping the datasets into two separate terms, period I (1991–1997) and period II (1998–2010), considering the difference in data acquisition patterns between the two study periods. In period I, the Cd data are available only as the annual average for each city. However, in period II, all measurement data were recorded consistently at monthly intervals at each station. As such, this period-based grouping scheme was used to evaluate the temporal patterns of Cd.

To make a parallel comparison of the data between the two periods and to analyze temporal trends over an extended period of 20 years, the monthly data in period II were first grouped together in an annual category to yield the respective annual mean values for each city. In addition, the seasonal patterns of Cd from seven individual cities were also evaluated by focusing mainly on the monthly Cd data obtained during period II.

RESULTS AND DISCUSSIONS

General Features of Cd Distributions in the Urban Air

As part of our efforts to learn more about the behavior of airborne Cd, its concentration levels were monitored concurrently along with those of other important metallic components over 20 years (1991–2010) period (Table 2). The Cd values generally varied in a comparable manner between different cities. If a comparison is made by the mean value of each city over the whole period, it changed moderately from 1.40 ng/m^3 (Daejeon) to 9.21 ng/m^3 (Ulsan). Hence, it those mean values are ranked by their magnitude, they are found in the following order: Ulsan > Incheon > Seoul > Busan > Daegu > Gwangju > Daejeon (Table 2).

As shown in Fig. 2, Cd values in most cities were much higher in period I than in period II. Interestingly, cities with low Cd levels (e.g., Busan, Daegu, Gwangju, and Daejeon) generally exhibited very little changes between the two periods. Ulsan and Incheon showed the highest Cd levels of all cities, and their annual means during period I generally exceeded the guideline value set by the European Union (EU) (5 ng/m^3). Such exceedance patterns from those 2 cities were also observed in the early part of period II. This guidance value was fixed by the Ministry of Environment, Heritage and Local Government in the EU (EU, 2009). They standardized the values of metals (including Cd) in ambient air to be observed in every European country (EU, 2009). As such, adequate information on the concentrations of metals has been made available to the public.

In Table 2, the Cd data measured at all the different cities are compared between the two time periods of I (1991–1997) and II (1998–2010), as explained above. Although temporal patterns of Cd changed considerably between the two periods, the highest values were seen consistently from Ulsan and Incheon, regardless of period.

Table 1. Basic information of the seven major metropolitan cities in Korea.

Order	Name of city	Area[a] (km^2)	Population[a] (million)	Population density[a] (km^{-2})	Annual average		Reference
					Temp. (°C)[b]	Humidity (%)[b]	
1	Seoul	605	9.8	16,000	8.6–17	64.4	Capital city
2	Busan	767	3.61	4,700	11.3–18.9	64.7	Largest port
3	Daegu	884	2.54	2,800	9.5–19.5	61.6	Fabric industry
4	Incheon	1,029	2.71	2,600	8.7–16.4	68.6	Diverse industrial facilities along the port
5	Gwangju	501	1.48	2,900	9.5–19.1	69.5	Large automobile company
6	Daejeon	540	1.54	2,800	8.3–18.4	66.7	Some minor industrial facilities
7	Ulsan	1,059	1.08	1,000	9.8–19.2	64.2	Heavy industry, ship building along the port

[a] Korean Statistical Information Service (Korean) as of 2 June 2010.
[b] Korea Meteorological Administration, 2010

Table 2. Comparison of the annual mean concentrations of Cd in the seven major cities in Korea throughout the entire study period 1991–2010.

Year	Target city of investigation (Cd(ng/m^3))[a]							Annual values for all cities[b]				
	Seoul	Busan	Daegu	Incheon	Gwangju	Daejeon	Ulsan	Mean	SD	CV	Min	Max
Period I (1991–1997)[c]												
1991	5.65	4.35	2.53	8.15	3.67		10.8	5.85	3.08	53	2.53	10.8
1992	9.08	2.02	2.08	19.70	2.07	0.63	9.45	6.43	6.87	107	0.63	19.7
1993	3.45	3.00	1.35	6.00	1.03	1.37	12.8	4.14	4.17	101	1.03	12.8
1994	3.50	2.85	2.13	6.66	1.15	1.73	11.3	4.18	3.60	86	1.15	11.3
1995	4.00	2.30	0.50	6.10	1.30	2.97	7.25	3.49	2.47	71	0.50	7.25
1996	3.10	2.23	3.18	6.25	1.40	2.00	16.8	4.99	5.42	109	1.40	16.8
1997	2.98	2.35	2.08	6.85	0.85	2.60	23.8	5.92	8.08	136	0.85	23.8
Period II (1998–2010)[d]												
1998	1.68	2.33	2.74	4.29	0.58	1.22	15.2	4.01	5.09	127	0.58	15.2
1999	1.73	3.35	1.77	4.78	0.68	1.80	16.8	4.42	5.63	128	0.68	16.8
2000	2.17	2.99	1.23	4.73	1.58	1.84	16.8	4.47	5.54	124	1.23	16.8
2001	2.98	2.46	2.19	6.43	1.42	1.38	11.6	4.07	3.74	92	1.38	11.6
2002	3.55	2.52	2.84	7.10	1.63	0.76	5.17	3.37	2.16	64	0.76	7.10
2003	2.55	2.01	2.28	9.94	1.63	1.05	4.39	3.41	3.06	90	1.05	9.94
2004	1.83	1.71	2.77	8.33	1.19	1.15	5.88	3.27	2.77	85	1.15	8.33
2005	1.19	1.94	2.20	3.79	2.29	1.18	3.35	2.28	1.00	44	1.18	3.79
2006	1.14	2.56	2.36	4.44	1.55	1.08	2.26	2.20	1.15	53	1.08	4.44
2007	1.23	2.14	2.61	3.94	2.29	1.18	2.66	2.29	0.94	41	1.18	3.94
2008	1.28	1.55	1.66	3.41	1.58	1.02	2.60	1.87	0.84	45	1.02	3.41
2009	1.35	1.68	1.27	2.08	0.88	0.71	2.81	1.54	0.73	47	0.71	2.81
2010	0.98	1.38	1.27	2.09	0.68	0.88	2.62	1.41	0.70	50	0.68	2.62
Period I (1991–1997)	4.54	2.73	1.98	8.53	1.64	1.88	13.1	4.92	4.36	89	1.64	13.1
Period II (1998–2010)	1.82	2.20	2.09	5.03	1.38	1.17	7.09	2.97	2.22	75	1.17	7.09

[a] Yearly mean value.
[b] SD and CV denote standard deviation and coefficient of variation, respectively.
[c] Period I: same as previous study of Kim (2007).
[d] Period II is evaluated newly in the present study.
[e] Results of t-test to check for the statistical significance of concentration differences between the study period of I and II are given as the probability (P) values.

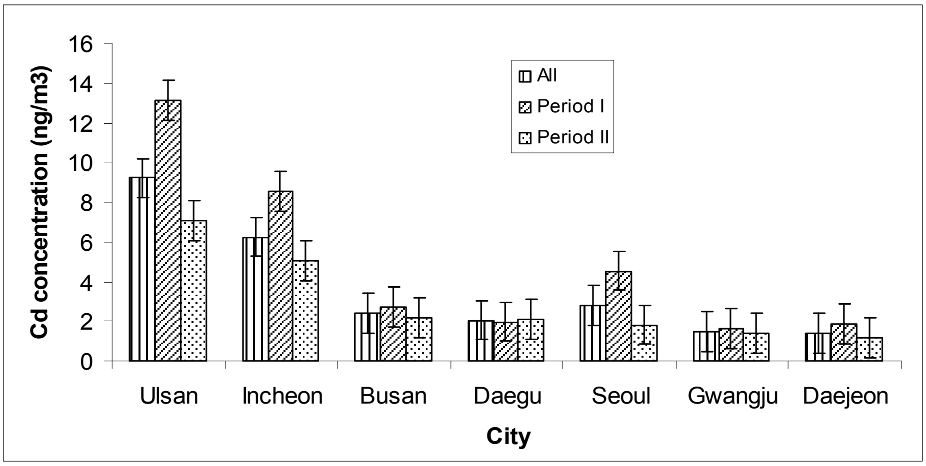


Fig. 2. Comparison of Cd concentration levels in seven cities between different temporal intervals: (a) all (1991–2010), (b) period I (1991–1997), and (c) period II (1998–2010).

In period I, the mean Cd values peaked in Ulsan (13.14 ± 5.53) and Incheon (8.53 ± 4.98). However, these values dropped to half of those levels during period II. As a result, the concentration levels of Cd during period II fell below European Union (EU) guideline value of 5 ng/m^3 in most cities except for Ulsan and Incheon. Such a reduction in Cd levels is likely to have been achieved by the combined effects of several factors including the adoption of new emission regulations, new manufacturing methods, and recycling of waste in the industries (e.g., Mutlu *et al.*, 2012). However, Cd levels in many countries still do not meet such guidelines despite considerable reductions achieved over the years (Mutlu *et al.*, 2012). For instance, relatively high concentrations of Cd were measured in some countries such as Nanling Mountains and the Pearl River Delta (urban zone), Southern China: 4.6–21.6 ng/m^3 during 2002–2003 (Lee *et al.*, 2005); Cartagena (urban zone), Spain: 2.9–8.9 ng/m^3 during 1991–1998 (Moreno-Grau *et al.*, 2000); and central Taiwan (urban area): 4.3 ng/m^3 during 2001–2002 (Fang *et al.*, 2003).

Seasonal Distribution of Airborne Cd

As the Cd data in period II were recorded on monthly basis, the results can be used to explore the seasonal patterns of Cd. Many studies have shown that the distribution of particulate matter (PM) is affected greatly by the seasonality of the meteorological conditions (Kim *et al.*, 2003, 2004; Kim, 2007). In Fig. 3, seasonal patterns of Cd in each city have also been evaluated (1) directly by the seasonal mean values and (2) after the normalization with respect to the overall mean values. To facilitate the comparison of Cd levels between seasons, the mean for each season at a given city was divided by the overall seasonal mean value of that city.

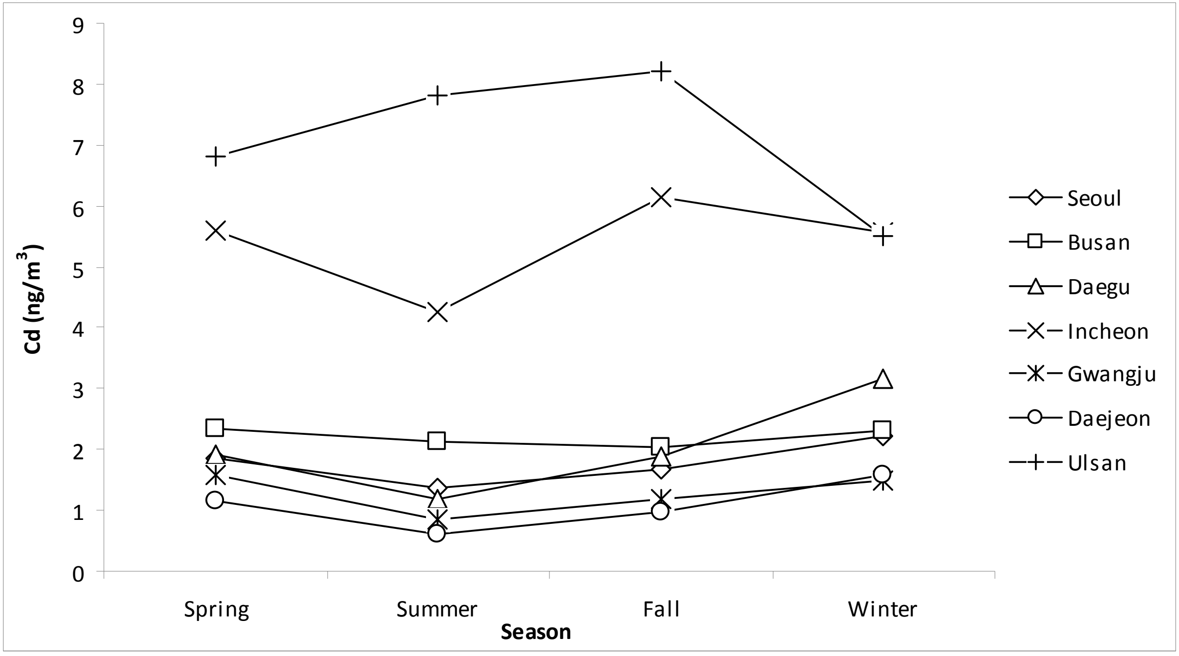
The seasonal trends of Cd data were similar in most target cities. Like the general pattern of PM data, the lowest Cd concentration occurred in summer, while the highest did so in winter. In contrast, the mean Cd values of Incheon were slightly higher in fall than in winter. Lim *et al.* (2010) reported that the PM concentration (including Cd) at a Daejeon industrial area increased in winter rather than other seasons, while observing a different seasonal trend in other industrial areas. As such, temporal patterns of Cd were often distinguishable between the most cities and ones with strong industrial activities (e.g., Ulsan). Although the data from these industrial (and port) areas like Ulsan tend to peak in summer (Basha *et al.*, 2007), the Cd values in Busan remained almost constant across seasons, showing little variation. The patterns observed in this study are slightly different from Mutlu *et al.* (2012) who disclosed relatively weak seasonal variation of Cd in most urban cities in Korea from 2004 to 2010. The observed differences may be explained by the selection of a more wider time band in this study (1998 to 2010) and slight differences in areal coverage (e.g., a total of 41 monitoring sites in this study vs. 30 monitoring sites for Mutlu *et al.* (2012)).

Seasonal patterns of Cd were also investigated at a roadside in Hong Kong from 2004–2005 (Cheng *et al.*, 2006; Shi *et al.*, 2012). According to these authors, the pattern of PM (including Cd) value increased dramatically in the winter season followed by spring and autumn, while the Cd values in summer showed a noticeable decreasing trend. However, there was very little deviation of PM values observed among the seasons (especially in summer and fall) as opposed to location. This similar trend was also observed in different countries in both industrial and residential areas. For instance, Cd values exhibited a greater increase in winter season than those of other seasons in Qingdao, China during 2001 to 2002 (Hao *et al.*, 2007), and Kolkata, India during 2003 to 2004 (Karar and Gupta, 2006). On the other hand, a different trend was observed with the highest values in fall and lowest in winter during 2006 to 2007 in Washington DC, USA (Melaku *et al.*, 2008).

Factors Controlling the Distribution of Cd

To investigate the factors controlling the behavior of Cd in the urban environment, a correlation analysis was conducted using the monthly Cd datasets measured between different cities in period II (e. g., the maximum of 156 monthly data points) for 13 years (1998–2010). This correlation analysis was conducted in two different respects: (1) correlations of

(A) Seasonal mean concentration of Cd



(B) Normalized seasonal mean values of Cd

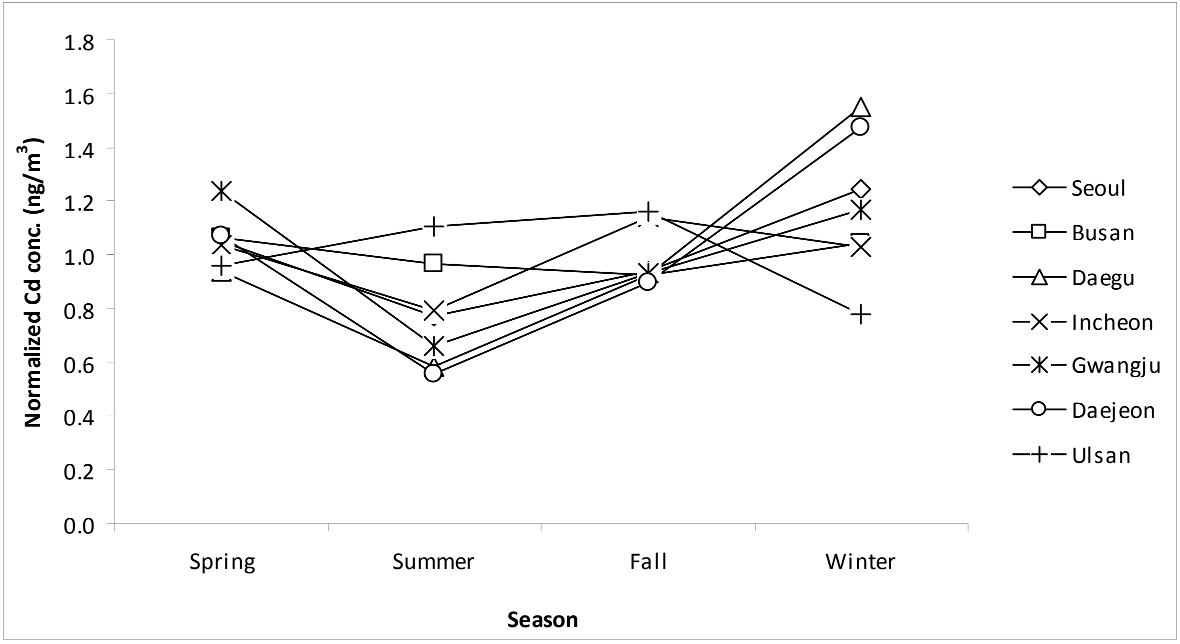


Fig. 3. Plot of seasonal mean values of Cd between seven cities in Korea during period II (1998–2010).

Cd data between different cities (Table 3(A)) and (2) correlations between Cd and other metal species for each city (Table 3(B)). In Table 3(A), the results of correlation analysis were highly comparable to show many matching pairs between different cities with statistical significance ($P < 0.01$). For instance, seven out of all 21 matching pairs showed the strongest correlations ($P < 0.01$), while there was only one pair with moderately strong correlation ($P < 0.05$) (Table 3(A)). The Cd data in Seoul generally showed fairly strong correlations ($p < 0.01$) with those of Busan, Daegue, and Daejeon. Likewise, the results of Daejeon showed significant correlations ($p < 0.01$) with those of Seoul, Busan, and Gwangju. Considering that different metals exhibit different correlation patterns in geographical sense, the observed patterns can imply a relatively good compatibility in the source properties of Cd in a fairly large geographical scale. Nonetheless, it may not necessarily be an essential component of Cd cycle.

In Table 3(B), the correlation analysis was also made to check the relationship between Cd and other metals in each city. The results indicate that there are abundant cases of significant correlations ($p < 0.01$) between most metals. The

Table 3. The results of correlation analysis using monthly Cd concentration data obtained from major urban areas in Korea during period II (1998–2010).

(A) Correlation of Cd data between different cities

City		Correlations						
		Seoul	Busan	Daegu	Incheon	Gwangju	Daejeon	Ulsan
Seoul	r	1						
	P							
	N	153						
Busan	r	0.284**	1					
	P	0.000						
	N	153	156					
Daegu	r	0.248**	0.133	1				
	P	0.002	0.101					
	N	149	152	152				
Incheon	r	0.139	0.014	0.043	1			
	P	0.086	0.861	0.601				
	N	153	155	151	155			
Gwangju	r	0.141	0.115	0.166	0.083	1		
	P	0.097	0.173	0.051	0.327			
	N	139	142	139	141	142		
Daejeon	r	0.302**	0.376**	0.207 *	0.110	0.285**	1	
	P	0.000	0.000	0.013	0.188	0.001		
	N	143	145	143	145	133	145	
Ulsan	r	0.118	0.323**	–0.081	0.057	–0.064	0.284**	1
	P	0.147	0.000	0.324	0.479	0.448	0.001	
	N	153	156	152	155	142	145	156

(B) Correlations between Cd and other metal species in each city

Pollutant		Seoul	Busan	Daegu	Incheon	Gwangju	Daejeon	Ulsan
Pb	r	0.533**	0.564**	0.631**	0.268**	0.620**	0.684**	0.257**
	P	1.26E-12	1.73E-14	3.04E-18	0.001	2.39E-16	2.57E-21	0.001
	N	153	156	152	156	141	145	155
Cr	r	0.200*	0.022	0.300**	0.135	0.024	0.272**	0.437**
	P	0.013	0.784	0	0.094	0.785	0.001	1.46E-08
	N	153	154	152	155	132	143	154
Cu	r	0.371**	0.425**	0.237**	0.208**	0.049	0.346**	0.534**
	P	2.31E-06	3.11E-08	0.003	0.009	0.565	1.98E-05	7.3E-13
	N	153	156	152	156	142	145	156
Mn	r	0.379**	0.315**	0.380**	0.262**	0.330**	0.447**	-0.085
	P	1.38E-06	6.3E-05	1.41E-06	0.001	6.43E-05	1.79E-08	0.291
	N	153	156	152	156	141	145	156
Fe	r	–0.134	–0.185*	–0.211**	–0.179*	–0.037	–0.118	–0.031
	P	0.099	0.021	0.009	0.026	0.66	0.158	0.698
	N	153	156	152	156	142	145	156
Ni	r	0.258**	0.263**	0.362**	0.257**	0.279**	0.221**	0.421**
	P	0.001	0.001	4.73E-06	0.001	0.001	0.008	4.55E-08
	N	152	156	152	156	142	145	156

** Correlation is significant at the 0.01 level (2-tailed).

* Correlation is significant at the 0.05 level (2-tailed).

strongest correlation ($p < 0.01$) of Cd was seen consistently with Pb and Ni from all seven cities. Although such strong correlations were also observed from Cu and Mn (from 6 cities), it was less abundant in the case of Cr (4 cities) and Fe (3 cities). This result thus suggests that the source properties of Cd should be tightly bound with other metals, although the level of such compatibilities may vary depending on the paired metal specie. It was reported that industrial and traffic emissions were the major contributors to the local airborne Cd (Basha *et al.*, 2010; Fang *et al.*, 2010). Thus, the high Cd concentrations in Ulsan and Incheon were likely to reflect the combined effects of local traffic and industrial emissions. Wu *et al.* (2007) also reported that vehicle exhaust was the main source of airborne metallic substances, including Cd. In contrast, average TSP concentrations decreased from the urban and industrial zones to residential areas. The same behavior is observed for Cd, Zn, and Pb, but not for Cu, which has a relatively short residence time (Morawska *et al.*, 2001; Espinosa *et al.*, 2002). It is interesting to note that Fe consistently maintained inverse correlations with Cd, regardless of statistical significance. According to Lee and Park (2010), Cd showed an inverse correlation with Fe in Ulsan during misty days in 2007. Similarly, Fe consistently showed inverse correlations with Pb and Cr. The observed patterns may suggest that a major crustal component like Fe exhibits behavior different from those made of an anthropogenic origin (like Cd). It is well known that Cd, Pb, and Cr generally come from various industrial activities (Pakkanen *et al.*, 2001; Lim *et al.*, 2010). One of the most important sources of heavy metals (including Cd) in the urban environment is also road traffic (Chandler, 1996; Ayras and Kashulina, 2000). Other contributors include waste incinerators, coal fired power plants, geogenic dust, and construction debris (Chandler, 1996; Ayras and Kashulina, 2000). The abundance and type of man-made activities can thus be a key factor leading to the alteration of Cd levels in urban and industrial area.

Long Term Variations in Cd Concentrations in Major Cities

Because the measurements of Cd had been made continuously in seven metropolitan cities in Korea over the past 20-year period, comparison of its annual mean values can be made to assess its long-term trend (Fig. 4). To conduct a long-term analysis, annual mean values of Cd obtained from all seven cities were examined by a simple linear regression analysis. The use of such simplified approach has commonly been made to provide a quick view of the general trend of many airborne pollutants (e.g., Hillery *et al.*, 1997). If the long-term trends of Cd concentrations are compared between cities, Cd concentrations in major metropolitan cities experienced gradual reductions throughout the past decades.

During the entire period (1991–2010), the Cd concentrations in all cities other than the two industrialized ones (Ulsan and Incheon) showed constant reductions across the years (Fig. 4(B)). In case of Ulsan, dramatic reduction was also seen from approximately 23.8 ng/m^3 (1997) to ~2 ng/m^3 (in recent years) (Fig. 4(A)). The Cd concentrations in Incheon exhibited a peak of 19.7 ng/m^3 in 1992, and then it declined continuously to approach ~2 ng/m^3 at present (Fig. 4(A)). The annual mean Cd values in Seoul showed a more than 40% reduction from 1998 (1.68 ± 0.61 ng/m^3) to 2010 (0.98 ± 0.39 ng/m^3) (Table 4). Likewise, other cities (Busan, Daegu, Incheon, and Daejeon) also showed similar reduction rates during period II (40.8, 53.7, 51.3, and 27.8%, respectively). However, as an exception, Gwangju with the one of the least Cd levels showed a slight increase from 0.58 ± 0.28 ng/m^3 (1998) to 0.68 ± 0.12 ng/m^3 (2010) (Table 4). In light of this complexity in long term trend of metals, a well-designed bio-monitoring plan is desirable for large scale, long term evaluations of environmental pollution caused by heavy metals (Dumchowski and Bytnerowicz, 2009).

A linear regression analysis of Cd was conducted to describe long-term trends of annual Cd data on a statistical basis (Fig. 5). The most significant trend ($P < 10^{-4}$) was seen in Seoul during the entire period (1991–2010), while the trends of Daegu and Gwangju exhibited the lowest significance ($P < 0.8$, and $P < 0.04$, respectively). On the other hand, Busan, Ulsan, Incheon, and Daejeon showed a moderately enhanced significance ($P < 0.01$). As such, most cities showed apparent reductions over the entire study period. A similar reduction pattern (~80%) was also observed in Europe during 1955–2005 (Pacyna *et al.*, 2009). In case of France, a reduction of 82% was seen from 1990 to 2006 (CITEPA, 2009). In addition, such a declining trend (~66%) was also seen in North-western Mediterranean, France from 1998 to 2008 (Heimbürger *et al.*, 2010). In Spain, the decreasing trend of Cd was 78% during 2001 to 2005 (Vicente *et al.*, 2012)

In industry, Cd can cause acute and chronic intoxications via inhalation and ingestion. Due to the long biologic half-life of Cd (10 to 40 years) in kidney (EC, 2007), it can exhibit a salient toxicological property in human body (Bernard, 2008). As a very small amount of Cd can cause greater deleterious effects than other toxic metals (Järup *et al.*, 1998; Bernard, 2004; Nordberg *et al.*, 2007), the US Environmental Protection Agency (USEPA) Integrated Risk Information System (IRIS) has recommended an inhalation unit risk (IUR) of Cd (as probable carcinogen) to be 1.8 ng/m^3 (Greene and Morris, 2006; Hieu and Lee, 2010). Accordingly, the results obtained in this study indicate some contrasting patterns between the city types. In 2010, Cd levels in most of the metropolitan cities (Seoul, Busan, Daegu, Gwangju, and Daejeon) of Korea remained in a relatively constant range of 0.68–1.38 ng/m^3. On the other hand, Cd levels in Ulsan and Incheon exhibited much higher values, above 2 ng/m^3 in recent years. The results in these industrialized cities can hence be of concern if one considers some of the health hazard guidelines.

CONCLUSIONS

In this study, Cd data were analyzed to evaluate its spatiotemporal variations in the major cities of Korea. The Cd concentrations measured at seven major cities were compared at different temporal intervals: (a) over the entire period of 20 years, (b) between years, and (c) between seasons. The Cd levels in highly industrialized areas exhibited the highest mean at 9.21 ng/m^3 (Ulsan) and 6.25 ng/m^3 (Incheon) for the 20 year period, while those of other urban areas showed moderately reduced values between 1.40–2.77 ng/m^3. The results suggest that the Cd concentration levels measured from the seven cities can be distinguished to a certain degree by the level of industrial activities.

To examine the temporal patterns of Cd, the data were analyzed between different seasons and across the entire study period. The seasonal patterns showed higher Cd concentration levels in winter in most cities. However, the Cd values at the highly industrialized city of Ulsan tend to peak in summer. When the mean Cd concentrations were analyzed on long-term basis, most cities consistently exhibited decreasing trends with statistical significance.

The results of this study confirm a constant reduction in

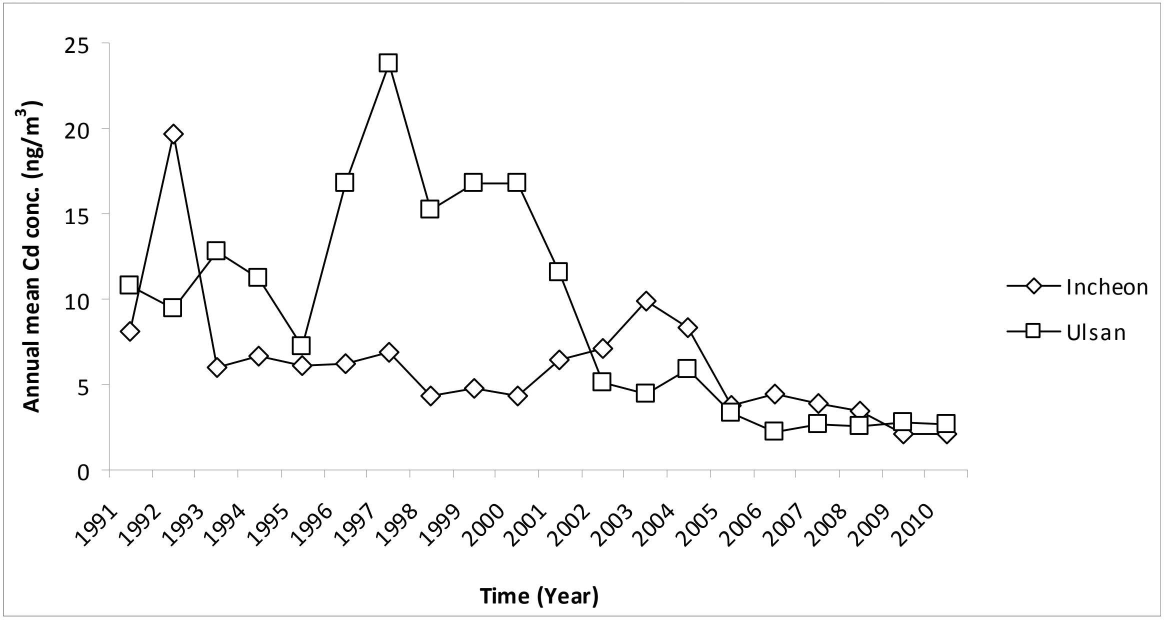
Table 4. Detailed summary of the Cd concentrations measured at monthly intervals from seven different metropolitan cities during period II (1998 to 2010).

Year	Cd (ng/m^3)						
	Seoul	Busan	Daegu	Incheon	Gwangju	Daejeon	Ulsan
All Year	1.82 ± 1.44 (1.40)[a] 0.10–7.8 (153(3))[b]	2.20 ± 1.01 (2.00) 0.10–5.6 (156)	2.09 ± 1.52 (1.60) 0.10–10.1 (152(4))	5.46 ± 6.47 (4.20) 0.10–68.80 (155(1))	1.40 ± 1.11 (1.20) 0.10–10.4 (142(14))	1.15 ± 0.81 (1.00) 0.10–3.90 (145(11))	7.09 ± 7.81 (3.85) 0.10–37.3 (156)
1998	1.68 ± 0.61 (1.70) 0.50–2.70 (12)	2.33 ± 1.08 (2.15) 0.80–4.30 (12)	2.74 ± 2.72 (1.60) 0.8–9.80 (11(1))	4.29 ± 2.31 (4.75) 0.90–8.60 (12)	0.58 ± 0.28 (0.70) 0.1–0.80 (5(7))	1.22 ± 0.84 (1.00) 0.3–3 (10(2))	15.23 ± 8.38 (15.45) 3.8–28.2 (12)
1999	1.73 ± 1.09 (2.1) 0.3–3.1 (12)	3.35 ± 1.02 (3.3) 2–5.6 (12)	1.77 ± 0.69 (2.00) 0.8–2.8 (9(3))	4.78 ± 4.08 (3.55) 0.1–11.6 (12)	0.68 ± 0.3 (0.75) 0.1–1.2 (12)	1.80 ± 0.96 (1.6) 0.3–3.3 (10(2))	16.83 ± 9.39 (14.35) 5.3–37.3 (12)
2000	2.16 ± 2.10 (1.45) 0.90–7.8 (10(2))	2.99 ± 0.99 (2.8) 1.4–4.9 (12)	1.23 ± 0.89 (1.45) 0.1–3.1 (12)	4.73 ± 2.14 (5.1) 1.3–7.5 (11(1))	1.58 ± 0.79 (1.6) 0.2–2.5 (11(1))	1.84 ± 0.84 (1.65) 0.6–3.5 (11(1))	16.76 ± 9.72 (14.6) 2.5–31.2 (12)
2001	2.98 ± 1.41 (3) 0.2–5.1 (12)	2.46 ± 0.52 (2.5) 1.7–3.2 (12)	2.19 ± 1.07 (1.8) 0.9–4.2 (12)	6.43 ± 2.41 (6.35) 2.7–10 (12)	1.42 ± 0.73 (1.5) 0.3–2.7 (12)	1.38 ± 1.00 (1.30) 0.2–3.7 (9(3))	11.61 ± 7.65 (13.6) 1.1–27.3 (12)
2002	3.55 ± 1.63 (3.3) 1.4–6.2 (12)	2.52–0.8 (2.5) 1.3–3.7 (12)	2.84 ± 1.16 (2.5) 1.4–4.4 (12)	7.1 ± 3.83 (5.95) 4–18.3 (12)	1.63 ± 0.68 (1.55) 0.5–3.1 (12)	0.76 ± 0.6 (0.6) 0.1–2 (12)	5.17 ± 6.77 (2.2) 0.3–23.3 (12)
2003	2.55 ± 2.23 (1.65) 0.1–6.9 (12)	2.01 ± 0.61 (1.95) 1.1–3.1 (12)	2.28 ± 0.87 (2.25) 1.2–4.1 (12)	9.94 ± 6.32 (7.75) 2.5–22.6 (12)	1.63 ± 0.56 (1.55) 0.9–2.5 (12)	1.05 ± 0.89 (0.70) 0.2–2.7 (11(1))	4.39 ± 4.73 (2.05) 0.8–17.3 (12)
2004	1.83 ± 1.90 (1.5) 0.2–6.8 (11(1))	1.71 ± 0.47 (1.64) 1.13–2.7 (12)	2.77 ± 1.39 (2.6) 1–5.1 (12)	8.33 ± 7.03 (6.25) 1.5–27 (12)	1.19 ± 1.01 (0.90) 0.1–3.6 (11(1))	1.15 ± 1.05 (0.9) 0.1–3.9 (12)	5.88 ± 3.74 (4.55) 2.2–13.8 (12)
2005	1.19 ± 0.71 (1.1) 0.4–3.11 (12)	1.94 ± 0.59 (1.85) 1–3 (12)	2.2 ± 1.56 (1.85) 0.7–6.7 (12)	3.79 ± 1.48 (4.06) 1.5–7.2 (12)	2.29 ± 3.18 (1.3) 0.2–10.4 (9(3))	1.18 ± 0.58 (1.10) 0.6–2.22 (10(2))	3.35 ± 2.22 (4.4) 0.4–6.3 (12)
2006	1.14 ± 0.57 (1) 0.4–2.2 (12)	2.56 ± 1.5 (2.85) 0.1–5.26 (12)	2.36 ± 1.56 (1.75) 0.7–5.7 (12)	4.44 ± 1.11 (4.69) 2.8–6.6 (12)	1.55 ± 0.55 (1.7) 0.7–2.3 (12)	1.08 ± 0.61 (1.2) 0.2–2.1 (12)	2.26 ± 1.36 (2) 0.5–6 (12)
2007	1.23 ± 0.98 (1) 0.2–3.4 (12)	2.14 ± 0.82 (2.05) 1.1–3.63 (12)	2.61 ± 2.64 (1.5) 0.7–10.1 (12)	3.94 ± 1.83 (3.9) 1.2–7 (12)	2.29 ± 1.01 (2) 1–4.5 (11(1))	1.18 ± 0.79 (1) 0.2–2.7 (12)	2.66 ± 1.02 (2.75) 1.1–4.4 (12)
2008	1.28 ± 0.56 (1.4) 0.5–2 (12)	1.55 ± 0.89 (1.4) 0.4–3.8 (12)	1.66 ± 1.18 (1.35) 0.4–4.8 (12)	3.41 ± 2.03 (3.05) 0.9–7 (12)	1.58 ± 0.76 (1.5) 0.8–3.2 (12)	1.02 ± 0.72 (1) 0.2–2.2 (12)	2.6 ± 1.45 (2) 0.6–5.1 (12)
2009	1.35 ± 0.6 (1.35) 0.5–2.7 (12)	1.68 ± 1.04 (1.4) 0.7–4.6 (12)	1.27 ± 0.41 (1.3) 0.7–2 (12)	2.08 ± 1.18 (1.75) 1–5.1 (12)	0.88 ± 0.62 (0.75) 0.4–2.8 (12)	0.71 ± 0.56 (0.55) 0.1–2.1 (12)	2.81 ± 2.88 (1.6) 0.2–8.8 (12)
2010	0.98 ± 0.39 (1.1) 0.4–1.5 (12)	1.38 ± 0.34 (1.25) 1–2 (12)	1.27 ± 0.73 (1.1) 0.4–2.8 (12)	2.09 ± 0.64 (1.9) 1.2–3.4 (12)	0.68 ± 0.12 (0.7) 0.4–0.9 (12)	0.88 ± 0.4 (0.85) 0.3–1.6 (12)	2.62 ± 2.84 (1.55) 0.1–9.4 (12)
Ratio (1998/2010)	1.71	1.69	2.16	2.05	0.85	1.39	5.81

[a] Mean ± SD (Median)

[b] Range: Min–Max (N); Here, N represents the number of monthly measurements.

(A) The two cities with the maximum Cd concentration levels (Ulsan and Incheon)



(B) The annual Cd values of the other five cities (Seoul and others)

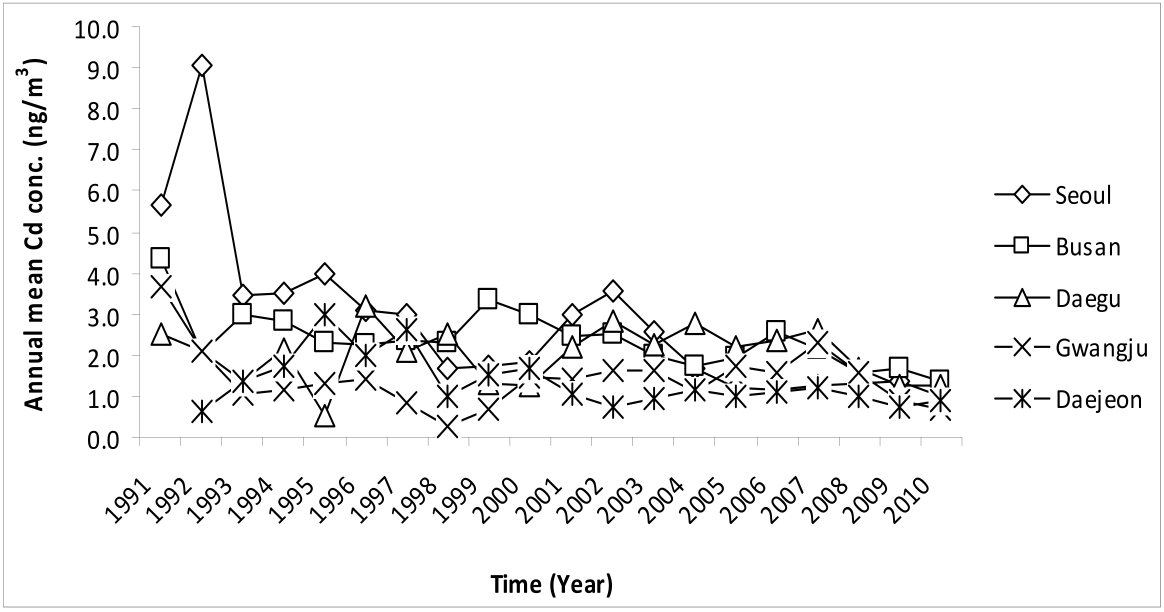


Fig. 4. Annual trend of Cd concentrations measured from all seven cities during the entire study period (1991–2010).

Cd values in all seven major cities during most of the study period. This indicates that the efforts of the Korean government to control anthropogenic emissions of Cd have been effective in a broad sense. However, Cd concentrations in certain cities such as Ulsan and Incheon still remain high enough, exceeding ~2 ng/m^3. Now, it is well known that Cd-containing products (batteries, alloys, coatings, pigments, etc.) are recyclable to a large extent and that the costs involved in such processes are in the economically acceptable range. The future studies are thus needed to develop more effective control methods and to tighten the individual anthropogenic emission sources of Cd along with other hazardous metal species. If such efforts are made successfully, the present level of Cd pollution observed in many industrialized areas can be reduced further.

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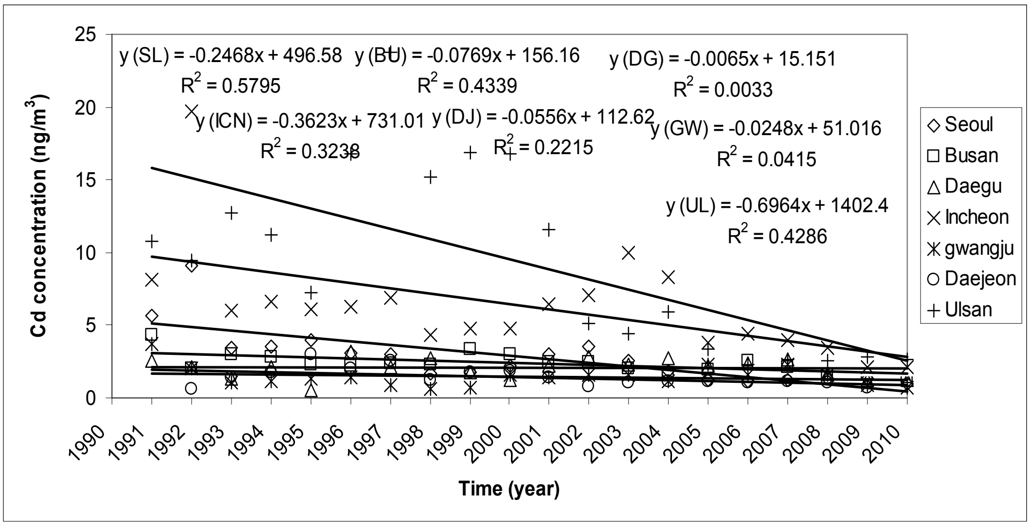

City	RF	R^2	Intercept	P
Seoul	–0.247	0.58	496.58	< .000
Busan	–0.077	0.434	156.16	< .002
Daegu	–0.007	0.003	15.15	< .8
Incheon	–0.326	0.324	731.01	< .009
Gwangju	–0.025	0.042	51.02	< .4
Daejeon	–0.056	0.222	112.62	< .03
Ulsan	–0.696	0.429	1402.4	< .002

Fig. 5. Linear regression analysis of the annual mean Cd values from all seven cities during the entire period (1991–2010).

REFERENCES

Aas, W., Alleman, L.Y., Bieber, E., Gladtke, D., Houdret, J., Karlssone, V. and Monies, C. (2009). Comparison of Methods for Measuring Atmospheric Deposition of Arsenic, Cadmium, Nickel and Lead. *J. Environ. Monit.* 11: 1276–1283.

ATSDR (Agency for Toxic Substances and Disease Registry) (2008). Cadmium Toxicity: Environmental Medicine and Educational Services Branch, US Department of Human and Health Services.

Avino, P., Capannesi, G. and Rosada, G. (2008). Heavy Metal Determination in Atmospheric Particulate Matter by Instrumental Neutron Activation Analysis. *Microchem. J.* 88: 97–106.

Ayras, M. and Kashulina, G. (2000). Regional Patterns of Element Contents in the Organic Horizon of Podzols in the Central Part of the Barents Region (Finland, Norway And Russia) with Special Reference to Heavy Metals (Co, Cr, Cu, Fe, Ni, Pb, V And Zn) and Sulphur as Indicators of Airborne Pollution. *J. Geochem. Explor.* 68: 127–144.

Basha, S., Gaur, P.M., Thorat, R.B., Trivedi, R.H., Mukhopadhyay, S.K., Anand, N., Desai, S.H., Mody, K.H. and Jha, B. (2007). Heavy Metal Content of Suspended Particulate Matter at World's Largest Ship-breaking Yard, Alang-Sosiya, India. *Water Air Soil Pollut.* 178: 373–384.

Basha, S., Jhala, J., Thorat, R., Goel, S., Trivedi, R., Shah, K., Gopalakrishnan, M., Gaur, P., Mody, K.H. and Jha, B. (2010). Assessment of Heavy Metal Content in Suspended Particulate Matter of Coastal Industrial Town, Mithapur, Gujarat, India. *Atmos. Res.* 97: 257–265.

Bernard, A. (2004). Renal Dysfunction Induced by Cadmium: Biomarkers of Critical Effects. *Biometals* 17: 519–23.

Bernard, A. (2008). Cadmium & Its Adverse Effects on Human Health. *Indian J. Med. Res.* 128: 557–564.

Bi, X., Feng, X., Yang, Y., Qiu, G., Li, G., Li, F., Liu, T., Fu, Z. and Jin, Z. (2006). Environmental Contamination of Heavy Metals from Zinc Smelting Areas in Hezhang County, Western Guizhou, China. *Environ. Int.* 32: 883–890.

Centre Inter Professionnel Technique d'Etudes de la Pollution Atmosphérique (CITEPA) (2009). Air Emissions in France, Mainland France, Substances Relative to the Contamination by Heavy Metals, www.citepa.org/emissions/nationale/ML/Emissions_FRmt_MLEN.pdf.

Chandler, A.J. (1996). Characterising Cadmium in Municipal Solid Waste, Sources of Cadmium in the Environment, Inter-Organisation Programme for the Sound Management of Chemicals (IOMC), Organization for Economic Co-operation and Development (OECD), Paris, France.

Cheng, Y., Ho, K.F., Lee, S.C. and Law S.W. (2006). Seasonal and Diurnal Variations of $PM_{1.0}$, $PM_{2.5}$ and PM_{10} in the Roadside Environment of Hong Kong. *China Part.* 4: 312–315.

Dmuchowski, W. and Bytnerowicz, A. (2009). Long-Term (1992–2004) Record of Lead, Cadmium, and Zinc Air Contamination in Warsaw, Poland: Determination by Chemical Analysis of Moss Bags and Leaves of Crimean Linden. *Environ. Pollut.* 157: 3413–3421.

EC (2007). European Union Risk Assessment Report. Cadmium Metal, Part II Human Health, 3rd Priority List, Volume 74, EUR 22767 EN, European Commission – Joint Research Centre, European Chemicals Bureau. Luxembourg: Office for Official Publications of the European Communities. Available at: http://ecb.jrc.it/documents/ExistingChemicals/RISK_ASSESSMENT/REPORT/cdmetalreport303.pdf.

Espinosa, A.J.F., Rodriguez, M.T., Rosa, F.J.B.D.L. and Sanchez, J.C.J. (2002). A Chemical Speciation of Trace Metals for Fine Urban Particles. *Atmos. Environ.* 36: 773–780.

EU (European Union) (2009). Arsenic, Cadmium, Mercury, Nickel and Polycyclic Aromatic Hydrocarbons in Ambient Air Regulations, Statutory Instruments, Stationery Office, Dublin, Ireland.

Fang, G.C., Chang, C.N., Chu, C.C., Wu, Y.S., Fu, P.P.C., Yang, I.L. and Chen, M.H. (2003). Characterization of Particulate, Metallic Elements of TSP, $PM_{2.5}$ and $PM_{2.5–10}$ Aerosols at a Farm Sampling Site in Taiwan, Taichung. *Sci. Total. Environ.* 308: 157–166.

Fang, G.C., Huang, Y.L. and Huang, J.H. (2010). Satudy of Atmospheric Metallic Elements in Asia during 2000-2007. *J. Hazard. Mater.* 180: 115–121.

Glorennec, P., Zmirou, D. and Bard, D. (2005). Public Health Benefits of Compliance with Current E.U. Emissions Standards for Municipal Waste Incinerators: A Health Risk Assessment with the Caltox Multimedia Exposure Model. *Environ. Int.* 31: 693–701.

Greene, N.A. and Morris, V.R. (2006). Assessment of Public Health Risks Associated with Atmospheric Exposure to $PM_{2.5}$ in Washington, DC, USA. *Int. J. Environ. Res. Public Health* 3: 86–97.

Hao, Y., Guo, Z., Yang, Z., Fang, M. and Feng, J. (2007). Seasonal Variations and Sources of Various Elements in the Atmospheric Aerosols in Qingdao, China. *Atmos. Res.* 85: 27–37.

Harmens, H., Norris, D.A., Koerber, G.R., Buse, A., Steinnes, E. and Ruhling, A. (2007). Temporal Trends in the Concentration of Arsenic, Chromium, Copper, Iron, Nickel, Vanadium and Zinc in Mosses across Europe between 1990 and 2000. *Atmos. Environ.* 41: 6673–6687.

Heimbürger, L.E., Migon, C., Dufour, A., Chiffoleau, J.F., and Cossa, D., (2010). Trace Metal Concentrations in the North-Western Mediterranean Atmospheric Aerosol between 1986 and 2008: Seasonal Patterns and Decadal Trends. *Sci. Total Environ.* 408: 2629–2638.

Hieu, N.T. and Lee, B.K. (2010). Characteristics of Particulate Matter and Metals in the Ambient Air from a Residential Area in the Largest Industrial City in Korea. *Atmos. Res.* 98: 526–537.

Hillery, B.R., Basu, I., Sweet, C.W. and Hites, R.A. (1997). Temporal and Spatial Trends in a Long-Term Study of Gas-Phase PCB Concentrations near the Great Lakes. *Environ. Sci. Technol.* 31: 1811–1816.

Järup, L., Berglund, M., Elinder, C.G., Nordberg, G. and Vahter, M. (1998). Health Effects of Cadmium Exposure - A Review of the Literature and a Risk Estimate. *Scand. J. Work Environ. Health* 24: 1–51.

Karar, K. and Gupta, A.K. (2006). Seasonal Variations and Chemical Characterization of Ambient PM_{10} at Residential and Industrial Sites of an Urban Region of Kolkata (Calcutta), India. *Atmos. Res.* 81: 36–53.

Kim, K.H., Choi, G.H., Kang, C.H., Lee, J.H., Kim, J.Y. Youn, Y.H. and Lee, S.R. (2003). The Chemical Composition of Fine and Coarse Particles in Relation with the Asian Dust Events. *Atmos. Environ.* 37: 753–765.

Kim, K.H., Choi, B.J., Yun, S.T. and Hwang, S.J. (2004). Studies of Spatial and Temporal Distribution Characteristics of TSP Bound Trace Metals in Seoul, Korea. *Environ. Int.* 127: 323–333.

Kim, K.H. (2007). Airborne Cadmium in the Major Monitoring Locations in Korea between 1991 and 2004. *Atmos. Environ.* 41: 4380–4395.

Kim, K.H., Ma, CJ. and Okuda, T. (2009). An Analysis of Long-Term Changes in Airborne Toxic Metals in South Korea's Two Largest Cities from 1991 to 2004. *Environ. Sci. Pollut. Res. Int.* 16: 565–572.

Lee, B.K. and Park, G.H. (2010). Characteristics of Heavy Metals in Airborne Particulate Matter on Misty and Clear Days. *J. Hazard. Mater.* 184: 406–416.

Lee, C.S.L., Li, X., Zhang, G., Peng, X. and Zhang, L. (2005). Biomonitoring of Trace Metals in the Atmosphere Using Moss (*Hypnum plumaeforme*) in the Nanling Mountains and the Pearl River Delta, Southern China. *Atmos. Environ.* 39: 397–407.

Lim, J.M., Lee, J.H., Moon, J.H. and Chung, Y.S. (2010). Source Apportionment of PM_{10} at a Small Industrial Area Using Positive Matrix Factorization. *Atmos. Res.* 95: 88–100.

Ma, C.J., Kang, G.U. and Kim, K.H. (2012). High loading for air pollution in the Byunsan Peninsula of Korea by an Interplay of the Saemangeum Project and winter Monsoon. *Asian J. Atmos. Environ.* 6–3: 234–243.

Melaku, S., Morris, V., Raghavan, D. and Hosten, C. (2008). Seasonal Variation of Heavy Metals in Ambient Air and Precipitation at a Single Site in Washington, DC. *Environ. Pollut.* 155: 88–98.

Morawska, L., Congrong, H., Hitchins, J., Gilbert, D. and Parappukkaran, S. (2001). The Relationship between Indoor and Outdoor Airborne Particles in the Residential Environment. *Atmos. Environ.* 35: 3463–3473.

Moreno-Grau, S., Perez-Tornell, A., Bayo, J., Moreno, J., Angosto, J.M. And Clavel, J.M. (2000). Particulate Matter and Heavy Metals in the Atmospheric Aerosol from Cartagena, Spain. *Atmos. Environ.* 34: 5161–5167.

Mutlu, A., Lee, B.K., Park, G.H., Yu, B.G. and Lee, C.H. (2012). Long-term Concentrations of Airborne Cadmium in Metropolitan Cities in Korea and Potential Health Risks. *Atmos. Environ.* 47: 164–173.

Nordberg, G., Nogawa, K., Nordberg, M. and Friberg, L. (2007). Cadmium, In *Handbook on Toxicology of Metals*, Nordberg, G., Fowler, B., Nordberg, M. and Friberg, L. (Eds.), Academic Press, New York, p. 65–78.

Pacyna, J.M., Pacyna, E.G. and Aas, W. (2009). Changes of Emissions and Atmospheric Deposition of Mercury, Lead, and Cadmium. *Atmos. Environ.* 43: 117–127.

Pakkanen, T., Loukkola, K., Kohonen, C., Aurela, M., Mäkelä, T., Hillamo, R., Aarnio, P., Koskentalo, T., Kousa, A. and Maenhaut, W. (2001). Sources and Chemical Composition of Atmospheric Fine and Coarse Particles in the Helsinki Area. *Atmos. Environ.* 35: 5381–5391.

Ray, S., Khillare, P.S. and Kim, K.H. (2012). The Effect of Aircraft Traffic Emissions on the Soil Surface Contamination Analysis around the International Airport in Delhi, India. *Asian J. Atmos. Environ.* 6–2: 118–126.

Shi, W., Wong, M.S., Wang, J. and Zhao, Y. (2012). Analysis of Airborne Particulate Matter ($PM_{2.5}$) over Hong Kong Using Remote Sensing and GIS. *Sensors* 12: 6825–6836

UNEP (United Nations Environment Programme) (2010). Final Review of Scientific Information on Cadmium Nairobi, Kenya.

USEPA (US Environmental Protection Agency) (2000). Cadmium Compounds. Technology Transfer Network, http://www.epa.gov/ttn/atw/hlthef/cadmium.html.

USEPA (US Environemental Protection Agency) (2011). Integrated Risk Information System, IRIS Substance List for Cadmium. http://www.epa.gov/iris/subst/0141.htm#suminhal.

Vicente, A.B., Sanfeliu, T. and Jordan, M.M. (2012). Assessment of PM_{10} Pollution Episodes in a Ceramic Cluster (NE Spain): Proposal of a New Quality Index for PM_{10}, As, Cd, Ni and Pb. *J. Environ. Manage.* 108: 92–101.

Wu, Y.S., Fang, G.C., Lee, W.J., Lee, J.F., Chang, C.C. and Lee, C.Z. (2007). A Review of Atmospheric Fine Particulate Matter and Its Associated Trace Metal Pollutants in Asian Countries during the Period 1995-2005. *J. Hazard. Mater.* 43: 511–515.

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