Journal of Hazardous Materials 188 (2011) 235–246

Contents lists available at ScienceDirect

Journal of Hazardous Materials

journal homepage: www.elsevier.com/locate/jhazmat

Nationwide shift in CO concentration levels in urban areas of Korea after 2000

Ki-Hyun Kim[a], Zang-Ho Shon[b,*]

[a] Department of Environment & Energy, Sejong University, Seoul 143-747, Republic of Korea
[b] Department of Environmental Engineering, Dong-Eui University, 995 Eom Gwang No, Busan Jin Gu, Busan 614-714, Republic of Korea

ARTICLE INFO

Article history:
Received 20 November 2010
Received in revised form 22 January 2011
Accepted 25 January 2011
Available online 1 February 2011

Keywords:
CO
Trend
Roadside
Mann–Kendall
Natural gas vehicle

ABSTRACT

Concentrations of carbon monoxide (CO) in urban and rural air were analyzed from 16 urban roadside locations in the 7 major cities along with 5 background areas in Korea during an 11-year period (1998–2008). Because of noticeable changes in CO levels after 2000, temporal evaluation of its roadside data was carried out by grouping them into period I (1998–2000) and II (2001–2008). The mean CO values for all 16 roadside stations between the two study periods I and II were significantly different from each other (1.67 ± 0.31 ppm (I) vs. 0.95 ± 0.17 ppm (II)). This interperiod reduction in CO levels fell, if compared between different stations, in the range of 8.62–59.94% (mean = 39.8 ± 14.7%). The statistical analysis confirms that CO concentrations decreased very rapidly with the annual reduction rate of 0.093 ppm year^{-1} (9.8% year^{-1}). In contrast, in background areas such distinctions are no longer valid between the two periods. A line of evidence collected in this study thus suggests that the implementation of legal and technical support (e.g., upgrading of fuel quality and the natural gas vehicle supply program) should have been the effective driving forces leading to the gradual reduction in CO levels in roadside locations (10 out of 16 stations) on the peninsula.

1. Introduction

Traffic activity in particular road transport releases various air pollutants (e.g., nitrogen oxides (NO_x), carbon monoxide (CO), carbon dioxide (CO_2), volatile organic compounds (VOC) and particulate matter (PM)). Its impacts have been significant on the atmospheric composition, air quality, and ultimately climate change [1–3]. Because of their environmental significance, numerous efforts have been made to reduce the emissions of CO and other pollutants (NO_x and fine particles) with the aid of legislative regulations and technical support in industrialized countries. In developing countries however, air pollutant emissions have been growing rapidly, posing a potential threat to human health. Especially in megacities (i.e., exceeding 10 million inhabitants), air pollution is now considered one of the most important problems [4–7].

Until recent decades, the main air pollutants of concern were sulfur dioxide (SO_2) and total suspended particulates (TSP) in Asian megacities such as Beijing and Delhi (as well as Mexico City). These pollutants are well known to be produced mostly by coal burning or motor vehicle exhaust [6]. Motor vehicle exhaust also accounts for the majority of NO_x and CO emissions in the megacities [6]. The air quality in most megacities has been improving due to emission control measures, except for NO_2 [5,6]. Unlike the common reports made, long-term air quality monitoring in Asian megacities has not been reported frequently in the literature, especially pollutants like CO.

To properly evaluate the current status of air pollution in a country whether developed or developing, the effectiveness of control efforts, and the direction of air quality change, a better knowledge of spatio-temporal trends of air pollutants in the past, present, and future is a prerequisite along with histories of emission inventories [8]. In addition, as air pollution in urban areas is dominated by vehicular sources, control measures have been devised and brought into practice in a variety of ways. Of the many technical approaches, the use of engines fueled by emission controlled diesel or compressed natural gas (CNG) has been proposed as a highly cost-effective management option [9]. In fact, vehicles fueled with natural gas have been demonstrated to dramatically reduce emissions of major pollutants like CO, CO_2, and reactive hydrocarbons. The consumption of natural gas for such vehicles has been widespread and welcomed both in the United States and Europe since the 1960s. Indeed, there are currently more than 30,000 natural gas vehicles on U.S. roads and over 700,000 worldwide.

Our preliminary study conducted to examine CO and CH_4 levels from a single urban roadside (U-RS) and its reference location in Seoul, Korea clearly demonstrated a line of evidence that changes in CO levels have likely been affected by such administrative efforts as the Natural Gas Vehicle Supply program [10]. In this study, we attempted to intensively examine the general patterns of CO level changes in relation to the implementation of the air quality control program by expanding the target areas of study to most major

---

[*] Corresponding author. Tel.: +82 51 890 2078; fax: +82 51 890 2076.
E-mail address: zangho@deu.ac.kr (Z.-H. Shon).

0304-3894/$ – see front matter © 2011 Elsevier B.V. All rights reserved.
doi:10.1016/j.jhazmat.2011.01.099

Table 1
Summary of the basic statistical information of CO concentrations for each individual station acquired via continuous measurements.

Order	City or province	Station information		Study period (year–month)		CO (ppm)					
		Code	Full name	Start	End	Mean	Median	SD	Min	Max	N
A. Urban roadside (U-RS) station											
1	Seoul	DD	Dong Dae Mun	98.1	08.12	1.46	1.20	0.88	0.50	5.20	128
2	Seoul	SU	Seoul STN	98.1	08.12	1.42	1.10	0.95	0.60	5.70	121
3	Seoul	CG	Cheong Gye Cheon	98.1	08.12	0.99	0.80	0.46	0.30	2.10	127
4	Seoul	CY	Cheong Nyang Ni	98.1	08.12	1.32	1.40	0.42	0.30	2.30	130
5	Seoul	SC	Sin Chon	98.1	08.12	1.00	0.80	0.44	0.40	2.10	127
6	Seoul	YD	Yeong Deung Po	98.1	08.12	0.94	0.70	0.45	0.30	2.00	128
7	Seoul	SS	Sin Sa	98.1	08.12	1.22	1.20	0.30	0.60	1.97	129
8	Bu San	OC	On Cheon	98.1	08.12	1.25	1.10	0.62	0.50	4.20	128
9	Bu San	CR	Cho Ryang	99.11	08.12	0.74	0.70	0.27	0.30	2.22	86
10	Dae Gu	NS	Nam San	98.1	08.12	1.35	1.30	0.45	0.60	2.63	119
11	Dae Gu	PR	Pyeong Ri	98.7	08.12	1.13	1.00	0.52	0.40	3.02	96
12	In Cheon	SB	Seok Ba Wi	98.1	08.12	0.99	0.91	0.30	0.40	1.64	129
13	Gwang Ju	CP	Chi Pyung	98.1	08.12	0.94	0.90	0.31	0.10	1.90	125
14	Gwang Ju	UA	Un Am	99.8	08.12	1.17	1.10	0.44	0.20	3.20	86
15	Dae Jeon	DH	Dae Heung 2	98.1	08.12	1.09	1.00	0.50	0.30	3.11	129
16	Ul San	SJ	Sin Jeong	99.11	08.12	0.86	0.60	0.60	0.30	3.30	71

Order	Province	Station information		Study period (year–month)		CO (ppm)					
		Code	Full name	Start	End	Mean	Median	SD	Min	Max	*N*
B. Background/outskirts stations											
1	In Cheon	SM	Seok Mo Ri	98. 1	08. 12	0.53	0.50	0.19	0.20	1.30	132
2	Gyeong Buk	TH	Tae Ha	98. 1	08. 12	0.39	0.40	0.15	0.17	1.20	128
3	Gyeong Nam	JG	Jeo Gu Ri	99. 11	08. 12	0.50	0.50	0.13	0.20	0.90	102
4	Chung Nam	PD	Pa Do Ri	98. 1	08. 12	0.39	0.40	0.20	0.10	0.80	130
5	Je Ju	GS	Go San Ri	98. 11	08. 12	0.41	0.40	0.18	0.10	0.90	117

urban areas including the megacity of Seoul in Korea. CO concentration data measured from 16 U-RS stations (in 7 major cities) between 1998 and 2008 were analyzed in relation to the reference CO data collected from 5 background stations across Korea. We attempted to elucidate the fundamental factors that affect the spatio-temporal behavior of CO in response to air quality control efforts. In addition, the annual data sets of CO in major urban locations of Korea are analyzed here to extract long-term variation trends between different stations over a decadal period.

2. Materials and methods

The concentration data of criteria pollutants (including CO, NO_2, SO_2, and PM) in major urban locations have been collected routinely from a total of 30 U-RS stations dispersed across 7 major metropolitan cities along with a number of small-scale cities in Korea (Table 1). Likewise, their concentrations are also monitored from as many as 32 regional background stations. All of these monitoring tasks are conducted as part of a routine operation managed by the Korean Ministry of Environment (KMOE). To investigate the spatio-temporal changes in the airborne pollutant levels in the largest urban locations in Korea, the concentration data of CO collected from the 30 U-RS stations were initially examined. The results from 14 out of these 30 stations were excluded however from further consideration since their coverage periods were not long enough to allow evaluations prior to 2000 (Table 1A). In order to compare the current status of air pollution in all the U-RS stations, the results derived from 32 background stations were also examined by the same criteria. These background stations are generally selected by the remoteness from major source activities. These background stations are selected by considering relatively strict criteria such as distance (from man-made sources), area population size, and so on. However, because of the limitations in the data coverage period, only the data from 5 background stations were selected as reference stations and analyzed in an analogous manner (Table 1B).

According to the summary of this compilation (Fig. 1), the 16 selected U-RS stations include 7 for Seoul and 9 for the 6 other major cities (i.e., Busan, Daegu, Incheon, Daejeon, Gwangju, and Ulsan). As these study stations are exposed to varying degrees of traffic activities, the behavior of CO is likely directly affected by such source processes [10]. For instance, the SC station in Seoul is located on the ground (sampling height of 3.8 m) at a distance of 1 m from an 8-lane road (30 m width). In order to explain the eminent role of traffic activities, the CO concentration data from all 16 U-RS stations were investigated in relation to the coinciding data sets from 5 background stations.

Although all the CO data from all monitoring stations were initially routinely recorded at hourly intervals, they were stored at monthly intervals in a data management network system operated by the Korea Ministry of Environment [11]. The detailed analysis of the CO data was hence made using its monthly representative (mean) values throughout the study period. The acquisition of raw CO data was made with a CO analyzer unit (response time = 60 s), which belongs to a component of on-line air quality measurement equipment (Maxsam-series, Kimoto, Japan). According to the manufacturer's specifications, the minimum detectable sensitivity of CO corresponds to 0.05 ppm with a precision of 0.5–2%.

To learn more about the temporal variation such as seasonal effect on CO distribution, we conducted a Kruskal–Wallis one-way analysis of variance (ANOVA) by rank testing of seasonal data group: spring (March–May), summer (June–August), fall (September–November), and winter (December–February). This statistical method is a non-parametric approach for testing the equality of population medians among data groups, which is identical to ANOVA, i.e., replacement of the data with their rank. In addition, the annual trend of CO concentration data was assessed using a non-parametric statistical method of the Mann–Kendall test. For the Mann–Kendall test, period I was not considered in long-term trend analysis due to insufficient data.

As shown in Tables 1S and 2S (supplemental material: SM), the database for the CO emissions in target cities and their local districts were obtained from the national air pollutants

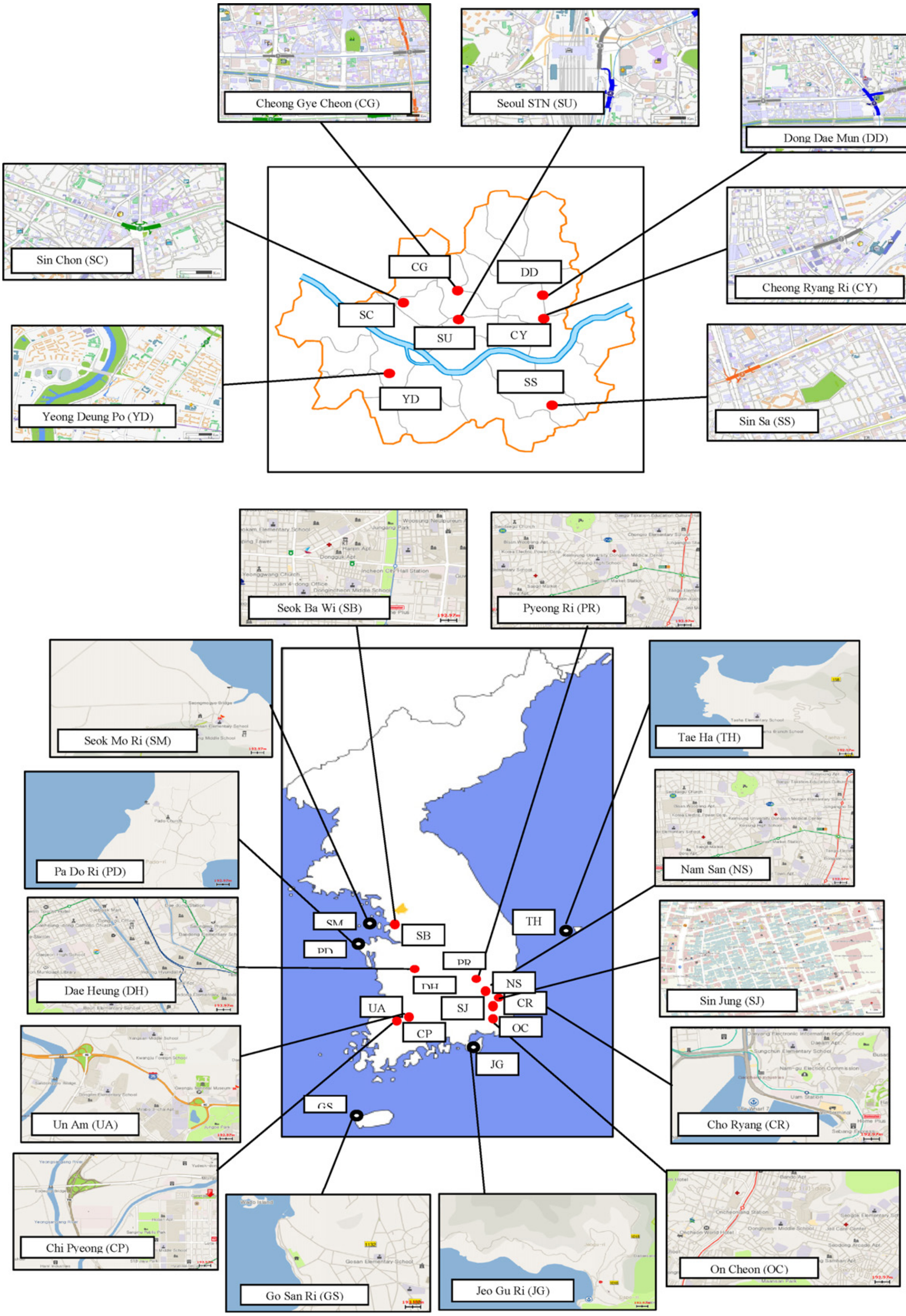

Fig. 1. Geographical locations of U-RS and background stations in Korea: (a) upper: 7 U-RS in Seoul (city location in orange color) and (b) lower: 9 U-RS (red) and 5 K-BG stations (black).

emission inventory system (CAPSS, clean air policy support system) by the National Institute of Environmental Research (http://airemiss.nier.go.kr/main.jsp) [12–20]. The yearly CO emissions were estimated on the basis of the source categories for the European Union emission inventory program [21] and national source categories [22]. Among several sources (point, area, mobile, fugitive and natural), the mobile (on-road) source is dominant (68% in 2007) [19]. The CO emissions from mobile sources were calculated using an emission factor and activity (total VKT, vehicle kilometers traveled with its average speed) (Tier 3 method). As the spatial scale of the CO emissions is available to cover the district level of each city, the emission magnitude for each station was approximated by the emission inventory database of the district scale. In other words, the emission value for a given station is assumed to be represented by the emission estimate for that local administrative district. For instance, as the district of Joong-Gu in Seoul city includes both DD and CG stations (see Tables 1S and 2S), both of them are assumed to have identical emission values.

3. Results and discussion

3.1. Spatial distribution of CO

In light of the noticeable changes in air quality after the implementation of natural gas vehicle supply policy in 2000 [10], all the data were examined after grouping them into two periods: I (January 1998–December 2000) and II (January 2001–December 2006). In Table 2, a statistical summary of the CO data is given to permit a comparison of two individual periods and their sum. The results of CO data for each U-RS station can be compared by the magnitude of their mean values derived for the entire study period. The mean values of all 16 U-RS stations during the entire period are computed as 1.15 ± 0.39 ppm (range of 0.74 (CR) to 1.46 ppm (DD)). These 2-fold differences in the mean CO levels between individual stations suggest that their distribution in the urban areas is likely to be affected by sources of fairly constant nature. Likewise, CO concentrations in 5 reference background stations also exhibit a relatively constant pattern but at a significantly reduced magnitude of 0.44 ± 0.10 ppm (in the range of 0.39 (TH and PD) to 0.53 ppm (SM)).

The spatial distribution of CO data is in general greatly distinguishable, if the comparison is confined to each individual period of I and II. Although differences in the mean values of U-RS and background stations were roughly five times during period I (1.67 vs. 0.36 ppm), the gap diminished greatly to 2-fold during period II (0.95 vs. 0.47 ppm). This comparison thus conforms to the relative dominance of traffic sources (U-RS) on CO distribution over other miscellaneous ones (background stations). The results of this comparison thus suggest that there is fairly good homogeneity in the CO distribution at stations of similar characters, as commonly seen from previous studies in various locations. It is however reasonable to infer that efforts to control air quality in urban areas have been the driving forces to reduce the spatial gaps in CO distribution.

3.2. Shift in CO concentration levels in urban area between before and after 2000

In light of our data acquisition interval, the general trends of the CO data in this study can be examined by three temporal criteria of month, year, and period. A brief inspection of the CO data indicates that the results share unique properties depending on temporal scales. To simplify the comparison of CO level changes across all study periods, the monthly mean concentration data for CO derived from U-RS stations can be plotted as a function of time. The results of most stations (13 out of all 16 U-RS stations), shown in Fig. 2, indicate three types of temporal patterns: (1) gradual decreases through the years (stations like Fig. 2a); (2) no gradual change but a sudden shift to lower CO levels after 2000 (Fig. 2b and d); and (3) trivial changes through time (Fig. 2c, e, and f).

As can be deduced from this simple comparison of mean CO levels between two periods using the *t*-test, the long-term patterns of the CO data seem to maintain rather unique properties of their own between different stations. Nonetheless, it is reasonable to conclude that most of them have been subject to noticeable reductions in CO levels from period I to II (Table 2). When the results are compared between the two periods, the patterns are consistent enough to show the reduction at all 16 U-RS stations without a single exception (Fig. 3a and b). A simple comparison of these U-RS data sets shows that the reduction between these two periods was estimated to be 39.8 ± 14.7% on average (range of 8.62 (SB) to 59.9% (SU)). In contrast, the results derived from 5 background stations show contrasting patterns between the two periods instead (Fig. 3c). In order words, it was found among these 5 background stations that SM, TH, and JG stations showed the reduction of 5–12.5%, whereas PD and GS stations underwent an increase of 147–194% (from 0.16/0.17 ppm in period I to 0.47 ppm in period II).

As the reduction in mean CO levels is found ubiquitously at all U-RS stations, it is important to assess the statistical significance of such concentration changes. When differences in two periods are compared by binding the annual concentration data for a given period, the patterns differ between Seoul and other cities. The differences in CO concentrations between the two periods ($p \leq 0.05$) for all 7 stations in Seoul were statistically significant. This pattern is slightly more complicated in other cities. For instance, the differences in CO concentrations between the two periods were statistically significant at 5 (out of 9) stations in other cities. These differences in mean CO concentrations at all U-RS stations (other than Seoul) were 34.3 ± 11.3% (34.9 ± 5.7% for the 5 stations), while those for all U-RS stations in Seoul were 46.9 ± 16.2%, on average. This geographical difference in mean U-RS CO levels between Seoul and other cities implies that control efforts for CO reduction were more effective in the former than the latter. Although all U-RS stations showed reductions in mean CO values from period I to II, 4 out of 16 stations were not statistically significant. In contrast, the results of 5 background stations show that only 2 stations maintained the patterns that are statistically significant (PD and GS) both of which exhibited inclining trends (rather than declining) during the study period. If percent difference in mean CO concentrations is derived for 2 significant cases (i.e., PD and GS), it is −170.6 ± 38.83%. On the other hand, that for the 3 remaining ones (i.e., SM, TH, and JG) is 7.1 ± 3.7%, on average. It is thus doubtless to say that there were not any practical efforts to control CO levels in those clean locations.

As seen from our analysis, one may consider that the large drop in annual CO concentrations after 2000 should be the most prominent phenomenon across the whole study period (Fig. 2). This unique pattern in CO distribution may be explained as the consequence of nationwide efforts to control air quality. In our previous study focusing on the distribution of CO and CH_4 at a single U-RS station (SC) in the megacity of Seoul, we postulated the possibility that the natural gas vehicle supply program acted as a major driving force leading to such a reduction in a highly efficient manner [10]. As active measures to control the urban air quality, the Korean government launched the natural gas vehicle supply program in June 2000 (in partial effort to maintain clean air quality during the 2002 FIFA World Cup) through which a total of 20,000 diesel fuel buses had ultimately been replaced with the natural gas buses by 2007 [23]. The positive signals of this policy had earlier been detected in the long-term trend of CO and SO_2 between 1997 and 2002 from some urban roadside areas in metro Daegu, Korea [24].

3.3. Temporal distribution of CO in urban areas

Although temporal trends derivable from all 16 U-RS stations are diverse, all the results share strong similarities in that they all underwent similar drops in CO concentration levels between the two periods. Moreover, it is quite striking to find that the patterns of certain stations exhibited a sudden drop in CO levels shortly after 2000 (e.g., CG, OC, SC, YD, and PR) (see Fig. 2). To quantitatively

Table 2

Comparison of CO concentration (ppm) data for each station between different temporal groups (period I, II and their sum (all)).

Period	Year	U-RS(all)[a]	DD	SU	CG	CY	SC	YD	SS
A1. U-RS stations (RS (all) for $N = 16$) and seven stations in Seoul)									
All	1998–2008	1.15 ± 0.39(1.03)[c] 0.57–2.45(132)[d,e]	1.46 ± 0.88(1.20) 0.50–5.20(128)	1.42 ± 0.95(1.10) 0.60–5.70(121)	0.99 ± 0.46(0.80) 2.10–0.30(127)	1.32 ± 0.42(1.40) 0.30–2.30(130)	1.00 ± 0.44(0.80) 0.40–2.10(127)	0.94 ± 0.45(0.70) 0.30–2.00(128)	1.22 ± 0.30(1.20) 0.60–1.97(129)
Period I	1998–2000	1.67 ± 0.31(0.62) 1.17–2.45(36)	2.45 ± 1.04(2.20) 1.22–5.20(35)	2.46 ± 1.14(2.35) 1.03–5.70(36)	1.65 ± 0.20(1.62) 1.24–2.10(36)	1.52 ± 0.20(1.50) 1.20–2.10(36)	1.62 ± 0.17(1.60) 1.30–2.10(36)	1.56 ± 0.21(1.60) 1.01–2.00(36)	1.55 ± 0.20(1.50) 1.30–1.97(35)
Period II	2001–2008	0.95 ± 0.17(0.93) 0.57–1.44(96)	1.08 ± 0.39(0.90) 0.5–2.30(93)	0.98 ± 0.30(0.90) 0.60–2.70(85)	0.73 ± 0.20(0.70) 0.30–1.60(91)	1.24 ± 0.46(1.20) 0.30–2.30(94)	0.75 ± 0.20(0.70) 0.40–1.60(91)	0.69 ± 0.23(0.65) 0.30–1.80(92)	1.09 ± 0.23(1.10) 0.60–1.80(94)
Percent difference between 2 periods		43.07	55.94	59.94	55.66	18.27	53.32	55.58	29.29
Statistical significance (p)		0.01	0.03	0.05	0.00	0.03	0.00	0.00	0.00

Period	Year	OC	CR	NS	PR	CP	UA	SB	DH	SJ
A2. U-RS stations other than Seoul ($N = 9$)										
All	1998–2008	1.25 ± 0.62 (1.10) 0.50–4.20(128)	0.74 ± 0.27 (0.70) 0.30–2.22(86)	1.35 ± 0.45 (1.30) 0.60–2.63(119)	1.13 ± 0.52 (1.00) 0.40–3.02(96)	0.94 ± 0.31 (0.90) 0.10–1.90(125)	1.17 ± 0.44 (1.10) 0.20–3.20(86)	0.99 ± 0.30 (0.91) 0.40–1.64(129)	1.09 ± 0.50 (1.00) 0.30–3.11(129)	0.86 ± 0.60 (0.60) 0.30–3.30(71)
Period I	1998–2000	1.90 ± 0.80 (1.70) 0.86–4.20(35)	1.07 ± 0.47 (1.03) 0.52–2.22 (11)	1.79 ± 0.46 (1.72) 0.85–2.63 (36)	1.61 ± 0.67 (1.40) 0.65–3.02 (24)	1.19 ± 0.26 (1.21) 0.50–1.62 (33)	1.58 ± 0.75 (1.51) 0.57–3.20 (12)	1.05 ± 0.36 (1.15) 0.40–1.64(36)	1.57 ± 0.52 (1.50) 0.86–3.11(33)	1.43 ± 0.36 (1.44) 0.87–1.87(5)
Period II	2001–2008	1.01 ± 0.28 (1.00) 0.50–2.00(93)	0.69 ± 0.18 (0.70) 0.30–1.10(75)	1.15 ± 0.28 (1.20) 0.60–1.80(83)	0.97 ± 0.34 (0.90) 0.40–2.00(72)	0.85 ± 0.28 (0.80) 0.10–1.90(92)	1.10 ± 0.33 (1.10) 0.2–1.7(74)	0.96 ± 0.28 (0.90) 0.40–1.50(93)	0.93 ± 0.38 (0.80) 0.30–2.30(96)	0.82 ± 0.60 (0.60) 0.30–3.30(66)
Percent difference between 2 periods		46.78	35.48	35.45	39.70	28.24	30.06	8.62	41.09	43.03
Statistical significance (p)		0.11	0.36	0.00	0.00	0.01	0.00	0.62	0.00	0.15

Period	Year	Background St. (all)[b]	SM	TH	JG	PD	GS
B. Background stations and its 5 individual component							
All	1998–2008	0.44 ± 0.10(0.44) 0.25–0.74(132)	0.53 ± 0.19(0.50) 0.20–1.30(132)	0.39 ± 0.15(0.40) 0.17–1.20(128)	0.50 ± 0.13(0.50) 0.20–0.90(102)	0.39 ± 0.20(0.40) 0.10–0.80(130)	0.41 ± 0.18(0.40) 0.10–0.90(117)
Period I	1998–2000	0.36 ± 0.07(0.35) 0.25–0.50(36)	0.55 ± 0.16(0.50) 0.30–1.00(36)	0.40 ± 0.14(0.40) 0.17–0.70(35)	0.56 ± 0.10(0.60) 0.30–0.70(14)	0.16 ± 0.09(0.10) 0.10–0.50(34)	0.19 ± 0.09(0.20) 0.10–0.50(26)
Period II	2001–2008	0.47 ± 0.09(0.46) 0.26–0.74(96)	0.52 ± 0.20(0.50) 0.20–1.30(96)	0.38 ± 0.15(0.40) 0.20–1.20(93)	0.49 ± 0.14(0.50) 0.20–0.90(88)	0.47 ± 0.17(0.50) 0.10–0.80(96)	0.47 ± 0.14(0.50) 0.20–0.90(91)
Percent difference between 2 periods		−29.7	5.31	4.70	11.36	−98.06	−143.20
Statistical significance (t-test, p)		0.51	0.51	0.82	0.28	0.00	0.00

[a] U-RS (all) data are created by combining monthly data of both 7 (Seoul) and 9 (non-Seoul) U-RS stations.

[b] Background station data are created by combining monthly data of both 5 background stations.

[c] Median value

[d] The number of data.

[e] Range

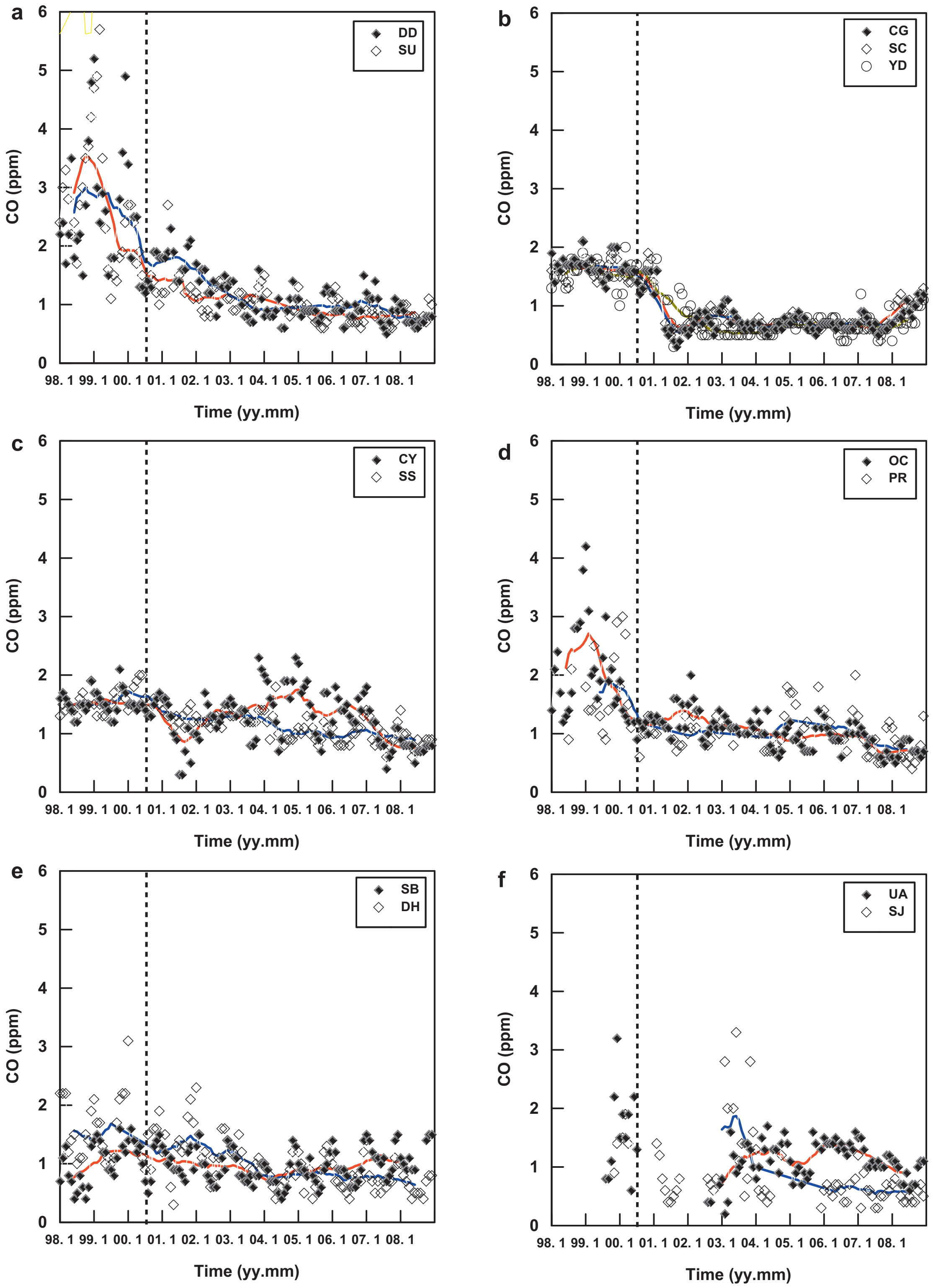

Fig. 2. Plot of CO concentration changes throughout all study period: the CO data from 13 out of 16 U-RS stations (7 U-RS stations in Seoul (a–c) and 6 U-RS stations in other urban areas) were used for this comparison. A 12 month moving average trend is fitted to the observation datasets.

assess the extent of CO level changes at all U-RS stations throughout the study period, the rate of reduction has been assessed along with its long-term trend for each station (Fig. 3).

In Table 3, the results of annual CO level change are compared between different stations using the data sets grouped as follows: (a) both periods, (b) period I, and (c) period II. In general, the long-term analysis of CO concentration data at 10 out of 16 U-RS stations shows clear downward trend at 95% confidence level ($p = 0.05$) according to the Mann–Kendall test. In contrast, the results derived at background stations did not show such a clear pattern. For both periods, the Sen's slopes of downward trend seen at U-RS stations ranged from 0.054 (SC) to 0.182 ppm year^{-1} (DD) with a mean slope of 0.093 ppm year^{-1}. For the calculation of statistical variables of CO trend and percent changes in Table 3, the stations with statistical

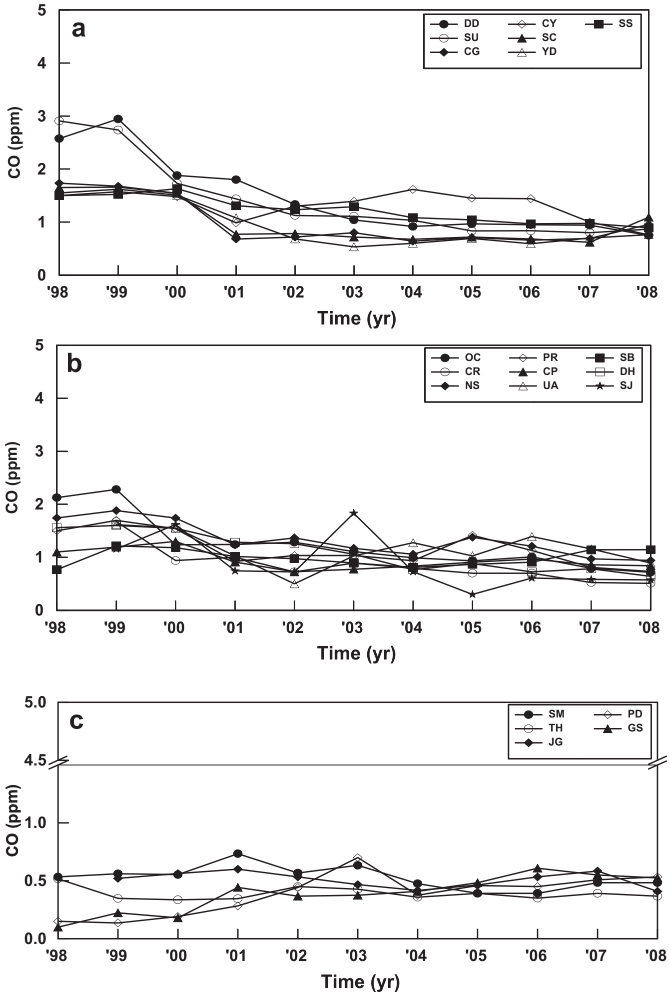

Fig. 3. Comparison of annual mean concentrations of CO across study period: (a) 7 U-RS in Seoul, (b) 9 U-RS in other urban areas, and (c) 5 K-BG stations.

significance were used. For period II, the downward trend (except for CG) was weakened by 35% on average, compared to that for both periods. In contrast, an upward (or a weak downward) trend of CO was prevalent during period I. These results imply that control efforts on CO emission became effective after 2000. For seasonal variability in CO distribution, 7 out of 16 U-RS stations showed patterns with statistical significance (Table 3). Although strong seasonality is apparent from many stations (e.g., CY, SB, and DH), it is not necessarily the case for the others (Table 3).

For CO concentration change rate, the mean for the annual reduction rate during the whole study period is computed to be 9.8% year^{-1}. The results of all sites other than SB show consistently declining trends in the range of 3.4 (CY) to 14% year^{-1} (SU). However, the results of the SB site are exceptional, exhibiting a slight gain such as 0.28% year^{-1}. However, note that this value fell within the error of the analysis and is statistically insignificant. When this approach is divided into the shorter time segments of periods I and II, the patterns are altered moderately to show either an increasing or decreasing pattern of annual CO change in several stations for each period. As the patterns of period I are reversed to show a slight gain from a few stations with statistical significance (SS, PR, and SJ), so do those of period II (SC only). In this respect, stations like both CR (68.4% year^{-1}) and SU (24% year^{-1}) showed significant reduction rates in period I. For period I, all stations in Seoul (except for SS) showed reduction, while stations in other urban areas recorded either clear gain (PR and SJ) or reduction (OC and CR). For period II, as most of the stations in Seoul experienced clear reduction (except for SC), so did those in other urban areas. The mean CO concentration changes for all stations with statistical significance (for the divided periods of I and II) are computed to be −9.6% and −6.3% year^{-1}, respectively. The results of this statistical analysis along with the comparison of mean CO levels between the two periods (I and II) consistently indicate that temporal trends of CO maintained strong consistency over a decadal period.

In Table 4, annual trends of ambient CO in large cities of the world are compared for the period between 1998 and 2008. Most of those cities showed statistically significant reduction in annual CO levels except for a few cities, e.g., Bucharest (Romania), Tallinn (Estonia), and Cairo (Egypt). The slope values for annual CO reduction ranged from 0.02 (Praha, Czech Republic) to 0.82 mg m^{-3} year^{-1} (Los Angeles). Note that relatively enhanced levels of CO at Los Angeles are representative of 8-h maximum values. The significant reduction in CO levels seen at Los Angeles might result from reinforced control measures. The reduction levels of annual CO in the cities of Paris (France), Athens (Greece), and Rome (Italy) in Europe (e.g., 0.15–0.20 mg m^{-3} year^{-1}) were somewhat higher than those seen at other cities in Europe. Meanwhile, some indirect means to control emissions such as the congestion charging scheme also functioned positively [1]. The congestion charging scheme in London, which was implemented in February 2003, led to the reduction of CO levels by up to 26% (between two years before and two after the introduction of the scheme).

In Asia, the CO reductions were significant in the megacities of Beijing (China, 0.14 mg m^{-3} year^{-1}) and Delhi (India, 0.30 mg m^{-3} year^{-1}). In Delhi, vehicles fueled with CNG and liquefied petroleum gas (LPG) have been demonstrated to induce dramatic reduction of pollutant emissions, as are the cases of CO, CO_2, reactive hydrocarbons, or NO_x [25,26]. As shown in Table 4, noticeable decreases in CO levels in Delhi after 2002 also reflect the effectiveness of its control efforts implemented since April 2001. However, their values seen in recent years (e.g., 1.4 and 2.2 mg m^{-3} in 2008, respectively) were still high (due to rapid population/automobile growth) relative to Europe and America. Annual reductions in ambient CO levels (e.g., 0.1 and 0.08 mg m^{-3} year^{-1}) in the cities of Seoul (this study) and Tokyo (Japan) were similar with relatively low CO levels (0.7–1.0 mg m^{-3}) compared to other Asian megacities. In addition, CO reductions in the megacities in Latin America (Mexico City (Mexico) and São Paulo (Brazil)) were also significant, but their recent CO levels (e.g., 2.3 and 1.9 mg m^{-3} in 2008, respectively) are still high. In Mexico City, changes in energy (fuel) use pattern such as a 14% increase in LPG fuel consumption from 1986 to 1999 has shown a noticeable effect on hydrocarbons and NO_x emissions [27], which might have a similar effect on CO levels. Detailed discussion on air quality in those Latin American megacities is given in [6]. In addition, the introduction of CNG in those Latin American megacities (Buenos Aires since 1995) has been beneficial in reducing the emissions of CO, hydrocarbons, and SO_2 [28]. Unlike other large cities, Cairo showed significant reduction in annual CO levels with a slope of 0.17 mg m^{-3} year^{-1}, which is statistically insignificant during the period of 2000–2006. The use of heavy-duty natural gas engines, in place of diesel, has also offered numerous environmental benefits in megacities [29].

3.4. Factors affecting CO concentration levels in urban areas of Korea

Fig. 4 depicts annual patterns for both CO concentration levels and CO emissions from natural gas vehicle (i.e., CO (CNG)) at each station. It also shows the percentage (%) of CO emissions due to CNG (CO (CNG)) relative to the total emission (i.e., CO (total)) for the station. The number of passenger cars (gasoline-powered in most cases) in Seoul, which is the major local source of CO in on-road mobile emission, increased from 1,710,000 to 2,225,000

Table 3
Comparison of temporal trends in CO concentration data across diverse temporal segments.

	Trend (ppm year^{-1})			Rate (% year^{-1})		
	All (slope, *p*)	Period II (slope, *p*)	Seasonal effect (*p*)	All	Period I	Period II
A. Roadside stations						
All				−8.38	−6.89	−4.28
DD	−0.182 (**0.0003**)	−0.097 (0.0003)	**0.020**	−13.7	−14.2	−10.5
SU	−0.151 (**0.0003**)	−0.076 (0.0003)	0.163	−14.0	−24.0	−8.02
CG	−0.057 (0.0873)	0.008 (0.0873)	0.136	−9.62	−5.82	2.34
CY	−0.032 (0.1188)	−0.020 (0.1188)	**0.000**	−3.36	−0.67	−2.97
SC	−0.054 (**0.0128**)	−0.018 (0.0128)	0.289	−8.87	−3.33	2.15
YD	−0.097 (0.1188)	0.004 (0.1188)	0.234	−10.9	−1.38	−3.18
SS	−0.067 (**0.0006**)	−0.062 (0.0006)	0.147	−5.90	4.19	−5.67
OC	−0.083 (**0.0011**)	−0.075 (0.0011)	0.051	−10.6	−23.4	−7.65
CR	−0.071 (**0.0009**)	−0.070 (0.0009)	0.708	−12.3	−68.4	−8.77
NS	−0.086 (**0.0031**)	−0.043 (0.0031)	**0.002**	−6.26	−0.04	−3.72
PR	−0.069 (**0.0349**)	−0.035 (0.0349)	**0.000**	−6.20	1.21	−2.09
CP	−0.026 (0.2757)	0.012 (0.2757)	**0.001**	−3.40	8.39	1.15
UA	−0.051 (0.3585)	0.058 (0.3585)	0.318	−3.55	−3.95	5.05
SB	−0.005 (1.0000)	0.020 (1.0000)	**0.000**	0.28	19.7	2.27
DH	−0.104 (**0.0003**)	−0.093 (0.0003)	**0.000**	−9.68	−0.38	−9.98
SJ	−0.060 (**0.0198**)	−0.027 (0.0198)	0.073	−10.9	32.1	−8.73
Average	−0.093	−0.060		−9.84	−9.63	−6.30
Median	−0.077	−0.066		−10.1	−1.86	−7.84
SD	0.042	0.028		3.03	26.1	4.00
Min	−0.054	−0.018		−14.0	−68.4	−10.5
Max	−0.182	−0.097		−5.90	32.1	2.15
B. Background stations						
All				−3.71	2.70	−7.58
SM	−0.010 (0.2113)	−0.040 (0.2113)	**0.001**	−3.13	1.77	−6.81
TH	0.000 (1.0000)	−0.006 (1.0000)	**0.010**	−1.23	−22.4	−1.07
JG	−0.012 (0.3222)	−0.013 (0.3222)	**0.000**	−1.75	7.51	−2.05
PD	0.043 (**0.0031**)	0.028 (0.0031)	0.222	10.5	13.2	3.61
GS	0.043 (**0.0019**)	0.030 (0.0019)	0.815	10.8	20.7	5.73
Average	0.043	0.029		10.7	17.0	4.67
Median	0.043	0.029		10.7	17.0	4.67
SD	0.000	0.001		0.21	5.30	1.50
Min	0.043	0.028		10.5	13.2	3.61
Max	0.043	0.030		10.8	20.7	5.73

Bold values are *p* values of less 0.05 (5%).

over the recent decade (2000–2008) (Table 1S). However, the CO emissions from gasoline-powered vehicles as well as total CO emission have decreased nationwide during that period (Fig. 4 and Table 2S). This CO emission reduction should be in part related to the improvement of fuel quality to counterbalance the increasing number of vehicles [30]. It was reported that CO emission from gasoline vehicles in 2006 was reduced by about 3%, compared to 2005 [31]. For all metropolitan cities (and other provinces), the yearly reduction rate of CO emissions from gasoline-powered vehicles was the largest from 2000 to 2001 with a mean reduction rate of 29% (Table 2S). Likewise, for all U-RS sites, its reduction rate from gasoline-powered vehicles was also the largest from 2000 to 2001 with a mean reduction rate of 20% (Table 2S).

Meanwhile, as shown in Fig. 4, shifts in CO concentration levels between before and after 2000 at most stations might also be affected to some extent by active clean-up efforts, i.e., the natural gas vehicle supply program. The replacement of vehicular fuel types was initiated from Seoul and 6 metropolitan cities in June 2000. This program was extended further to cover small-sized urban districts in 8 provinces in 2003. This active clean-up activity along with other miscellaneous administrative efforts has been effective enough to induce large changes in urban air quality, especially with respect to carbon monoxide. The largest increase in CNG-based emissions of CO among all cities (e.g., by two orders of magnitude) is found in Seoul (from 19 to 2185 ton year^{-1} over 7 years) (Table 2S). Although CO emitted by the consumption of CNG represents only a small proportion of its total emission, its relative proportion increased dramatically during the study period (e.g., from less than 0.01% (2000) to 1.53% (2007) in Seoul).

The results of correlation analysis between annual ambient CO concentration and its emissions (from gasoline and CNG vehicles) is given in Table 3S. The annual CO concentrations at all U-RS sites in Seoul were positively correlated with CO emissions from gasoline-powered vehicles with high correlation coefficients ($R > 0.78$, except for CY (0.41). However, its concentrations were inversely correlated with emissions from CNG buses ($R > 0.50$). At most U-RS sites in other cities, the annual CO levels were also strongly correlated with CO emissions from gasoline vehicles ($R > 0.72$, except for CP, UA, DH, and SJ). In contrast, at DH and SJ, their correlations showed opposite trends (i.e., inverse correlation), without statistical significance. Meanwhile, such correlation patterns for CNG buses at the U-RS sites were fairly different between Seoul and other cites. In other words, 6 out of 9 sites exhibited inverse correlations. This implies that CO emission control measures (such as replacement with CNG buses) at certain sites in other cities were not as effective as in Seoul.

Inter- and intra-seasonal variations in CO levels at U-RS and background stations are described in Fig. 5. There were significant differences in inter- and intra-seasonal variations depending on stations. The amplitudes (i.e., difference in CO levels between Max. (or Min.) and mean) of the CO variation at the U-RS stations ranged from 7.3 to 47.8% (mean of 23.4 ± 10.1%), while those at the background stations ranged from 11.7 to 25.8% (mean of 19.6 ± 4.4%). The difference between maximum and minimum CO values at the U-RS stations (e.g., 0.92 ppm) occurred at DD in Seoul, while that at the background station (0.24 ppm) at SM. The CO level variation at the U-RS stations was somewhat smaller in Seoul (18.7 ± 9.9%)

Table 4
Annual trends of ambient CO (1998–2008) in large cities of the world (including megacities).

City	Annual average CO concentrations ($mg\,m^{-3}$ (ppm))											Slope[m] ($mg\,m^{-3}\,year^{-1}$)
	1998	1999	2000	2001	2002	2003	2004	2005	2006	2007	2008	
Praha[a]	1.0	0.8	0.8	0.9	0.8	1.0	0.8	0.7	0.8	0.7	0.7	−0.02[*]
Bucharest[a]							1.7	1.4	2.2	1.5	0.8	−0.21
Berlin[a]		0.9	0.8	0.7	0.7	0.7	0.6	0.5	0.4	0.4	0.2	−0.07[***]
Bern[a]	1.2	1.1	1.1	1.0	1.0	0.7	0.6	0.7	0.6	0.5	0.5	−0.08[***]
Paris[b]	2.4	2.2	1.8	1.6	1.5	1.5	1.3	1.2	1.1	1.0	0.7	−0.15[***]
Mardrid[a]	1.9	1.2	1.1	1.0	0.7	0.7	0.7	0.6	0.6	0.5	0.4	−0.10[***]
Rome[a]	3.0	2.5	2.5	2.1	1.9	1.6	1.5	1.2	1.3	1.1	1.0	−0.20[***]
Athens[a]		1.7	1.3	3.0	2.9	2.5	2.5	2.3	2.2	2.0	1.8	−0.14(0.17[n,***])
London[a]	1.8	1.5	1.4	1.2	0.9	0.8	0.8	0.7	0.7	0.7	0.6	−0.10[***]
Stockholm[a]	1.4	1.3	1.0	0.9	0.8	0.8	0.7	0.6	0.5	0.5	0.4	−0.10[***]
Tallinn[a]								0.4	0.4	0.4	0.3	−0.02
Moscow[c]		1.3	1.4	1.1	1.7	1.4						0.06
Beijing[d]	3.3	2.9	2.7	2.6	2.5	2.4	2.2	2.0	2.1	2.0	1.4	−0.14[***]
Tokyo[e]	1.4(1.2)[l]	1.2(1.1)	1.3(1.1)	1.2(1.0)	1.1(0.9)	1.0(0.9)	0.9(0.8)	0.9(0.7)	0.8(0.7)	0.7(0.6)	0.7(0.6)	−0.08[***]
Seoul[o]	2.1(1.7)	2.1(1.8)	1.8(1.5)	1.3(1.1)	1.2(1.0)	1.2(1.0)	1.1(0.9)	1.1(0.9)	1.1(0.9)	1.0(0.8)	1.0(0.8)	−0.10[***]
Delhi[f]	5.5	4.2	4.7	4.2	3.3	2.8	2.6	2.5	2.5	2.5	2.2	−0.30[***]
New York[g]	2.0(1.7)	1.8(1.5)	1.6(1.4)	1.4(1.2)	1.2(1.1)	1.3(1.1)	1.1(1.0)	1.1(0.9)	1.0(0.8)	0.7(0.6)		−0.13[***]
Los Angeles[h]	11.5(9.8)	11.2(9.5)	9.6(8.2)	7.0(6.0)	7.1(6.1)	6.7(5.7)	5.7(4.9)	4.3(3.7)	4.1(3.5)	4.1(3.5)	3.8(3.2)	−0.82[***]
Mexico City[i]	4.8(4.1)	4.6(3.9)	3.6(3.1)	3.7(3.2)	3.5(3.0)	2.6(2.3)	1.6(1.4)	2.2(1.8)	2.1(1.8)	2.1(1.8)	2.3(1.9)	−0.28[**]
São Paulo[j]		3.0(2.6)	2.9(2.5)	2.9(2.5)	2.8(2.4)	2.5(2.1)	2.3(2.0)	2.5(2.1)	2.2(1.9)	2.2(1.9)	1.9(1.6)	−0.12[***]
Cairo[k]			4.4	8.2	5.0	5.4	6.8	5.3	3.4			−0.17

[a] AirBase – European air quality database (http://air-climate.eionet.europa.eu/databases/airbase/); Annual mean value of hourly CO; roadside stations.
[b] http://www.airparif.asso.fr/pages/resultats/histostats; Annual mean value of hourly CO; roadside stations.
[c] [36].
[d] [37].
[e] http://www.nies.go.jp/igreen/td_disp.html; roadside stations.
[f] Central Pollution Control Board (http://www.cpcb.nic.in/annual_archive.php); Annual mean value of 8-h CO; roadside stations.
[g] http://www.dec.ny.gov/chemical/8406.html (New York State Ambient Air Monitoring System); roadside stations.
[h] South Coast Air Quality Management District (http://www.aqmd.gov/smog/historicaldata.htm); Annual mean value of 8-h maximum values.
[i] http://www2.ine.gob.mx/dgicurg/calaire/tend/horarias/concentra.php?ciudad1=ZMVM; Annual mean value of hourly CO.
[j] http://www.cetesb.sp.gov.br/Ar/publicacoes.asp (Relatório de qualidade do ar no Estado de São Paulo); Annual mean value of daily 8-h maximum values.
[k] Egyptian Environmental Affairs Agency – Air Quality in Egypt, 2000 (http://www.eeaa.gov.e.g./English/main/achivements.asp).
[l] The number in parenthesis is CO concentration in ppm.
[m] Mann–Kendall test.
[o] This study.
[n] When data at Athens in 1999–2000 are excluded, the trend is statistically significant with slope of 0.17.
[*] p value: $0.001 < p < 0.01$.
[**] p value: $0.001 < p < 0.01$.
[***] p value: $0.001 < p$.

than that in other cites (27.0 ± 9.5%). Thus, the magnitudes of the inter- and intra-seasonal variations were slightly different between U-RS and background stations, with the enhanced variations at the former in part due probably to anthropogenic sources.

Although relative contribution of on-road mobile sources to the total budget of anthropogenic sources has decreased from 81.1 (1999) to 67.6% (2007), it is still the predominant source sector of CO emission [12–20]. If we assess the temporal pattern of countrywide energy (especially gasoline and diesel) consumption in the transportation sector, such information will allow us to deduce the comparable variation in CO emissions; this is because both of them are its major on-road mobile sources. The national energy consumption data sets were obtained from the Korean Energy Economics Institute (KEEI, http://www.keei.re.kr/main.nsf/index.html) [32]. As shown in Fig. 1S, there was no clear pattern in temporal distribution of energy consumption by gasoline and diesel. For instance, the maximum and minimum consumption in 2006 occurred during summer months, the former in June and the latter in July, respectively. In 2007, the maximum occurred in June, whereas the minimum in February; in 2008, the maximum occurred in December, whereas the minimum in July. The amplitude of inter- and intra-seasonal variations in energy consumption by transportation was relatively small (e.g., <20% for the sum of diesel and gasoline) compared to the magnitude of ambient CO variation. Hence, temporal variation (amplitude) of CO concentration might be affected less significantly by its emission from the on-road mobile sources than its loss processes.

Seasonal patterns of CO levels characterized by high winter (December or January) and low summer months (July or August) may be ascribed to the combined effects of anthropogenic sources (i.e., on-road mobile sources), meteorological conditions, and photochemical reactions. More specifically, low CO levels in summer are likely to result from such factors as strong physico-chemical removal (by intensive precipitation), the air mass pathways in long-range transport (i.e., less polluted air mass), its relatively enhanced photochemical loss by OH, and/or reduced emissions from on-road mobile CO sources. A summary of seasonal variations in major meteorological parameters is given for both Seoul (Table 5) and other cities (Table 4S). General features in meteorological parameters are comparable to each other. The precipitation intensity is significantly higher during summer (e.g., 387 mm in Seoul and 218–296 mm in other cities) than during winter (e.g., 20 mm in Seoul and 19–40 mm in other cities) which is different by an order of magnitude. In addition, temperature difference between summer and winter is significantly large in Seoul (24.5 °C) relative to other cities (18.5–23.6 °C); this effect can cause significant difference in photochemical loss reactivity of CO (by OH oxidation). Significant difference in OH concentrations between summer and winter (by approximately an order of magnitude)

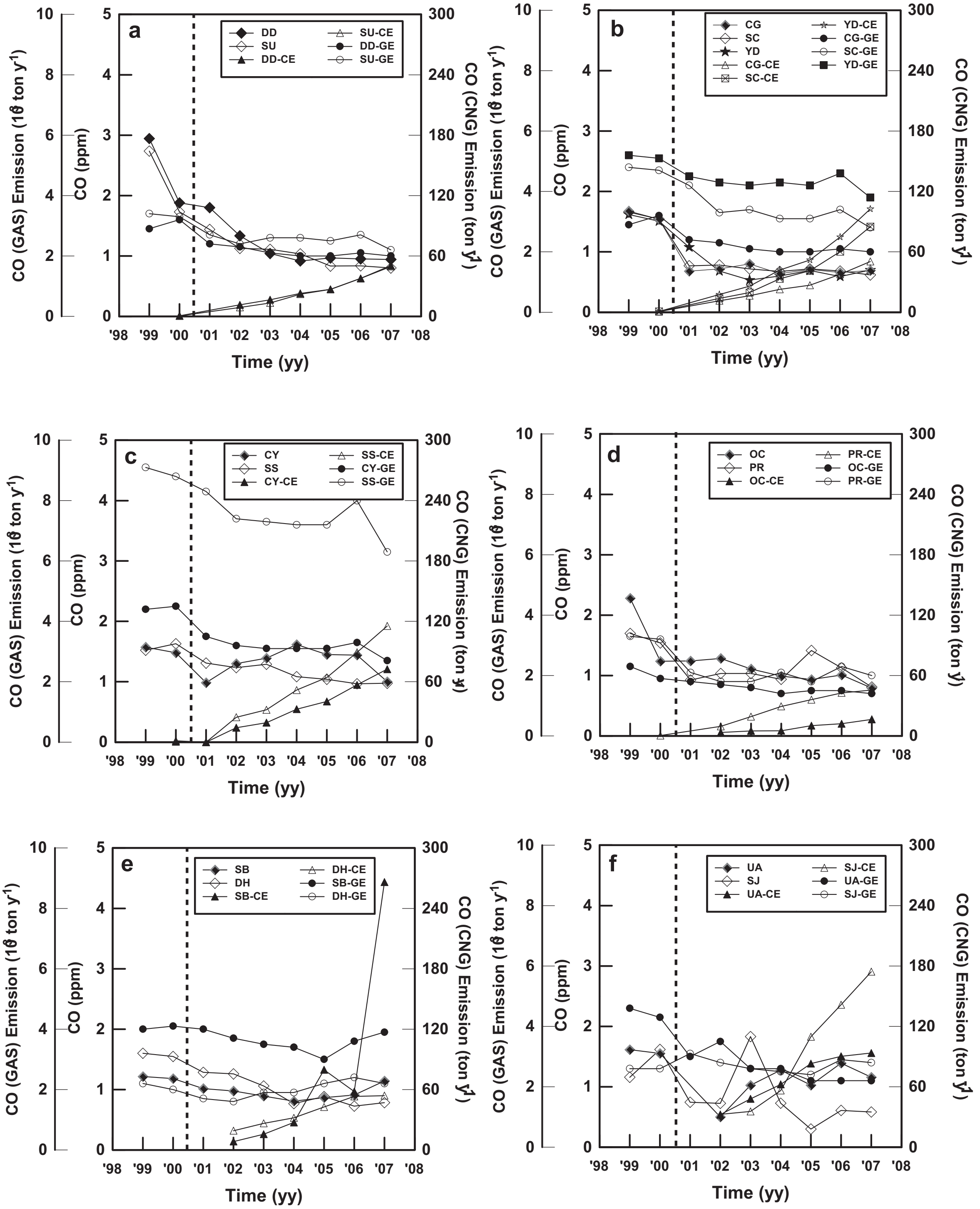

Fig. 4. Comparison of annual CO concentration and its emission from CNG buses (i.e., CO (CNG)) and gasoline vehicles (i.e., CO (GAS)) at each site. In legend box, the site symbols ending with -CE and -GE represent CO emissions due to CNG buses and gasoline vehicles, respectively.

[33] can also contribute at least partially to the seasonal difference in CO levels. In contrast, lower wind speed in summer (e.g., in Seoul $2.1 \pm 0.4\,\mathrm{m\,s^{-1}}$ in summer vs. $2.3 \pm 0.2\,\mathrm{m\,s^{-1}}$ in winter) which can cause moderately slow horizontal advection (dilution) does not seem to contribute to low CO levels in summer. In Seoul, easterly wind, ENE, was dominant in summer, while that of westerly wind, WNW, was dominant in winter. Wind rose patterns in other cities, where southerly and northerly winds were dominant in summer and winter, respectively, are somewhat different from that in Seoul. Long-range transport can also be considered as a potent source of CO in the study area, due to its relatively long lifetime (i.e., about 2 months [34]) and to the presence of a nearby major man-made source area (i.e., China). High CO level during winter season at U-RS sites in Seoul (and other cities) may be in part related to the long-range transport from China based on the analysis of dominant wind direction of WNW, westerly winds (and

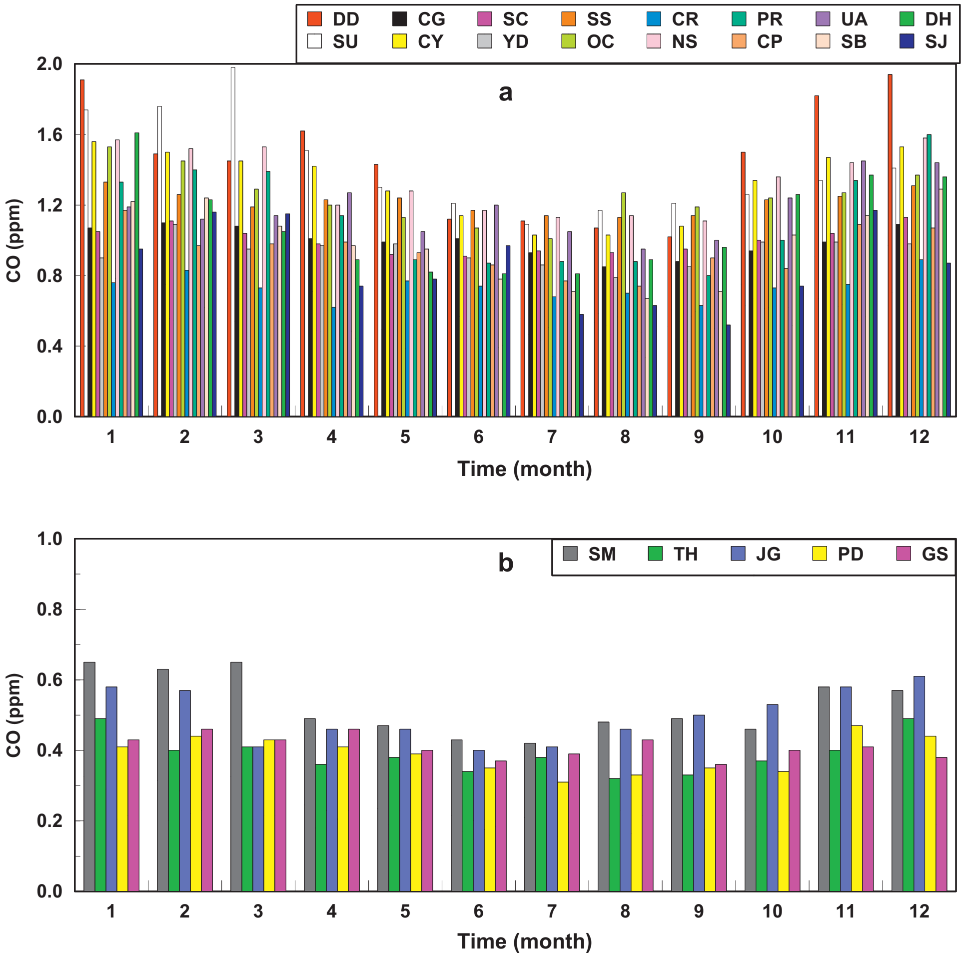

Fig. 5. Inter- and intra-seasonal comparison of CO concentration levels during study periods: (a) roadside stations and (b) background stations.

Table 5
Comparison of seasonal variation in meteorological parameters in Seoul during the study periods.

		Year											Average
		1998	1999	2000	2001	2002	2003	2004	2005	2006	2007	2008	
Temperature (°C)	Winter	1.3	0.7	−1.1	−1.2	0.9	0.6	0.5	−0.8	−1.3	1.9	−0.4	0.1 ± 1.1
	Spring	14.0	12.7	11.9	12.6	13.1	13.1	12.3	11.7	11.7	11.9	13.0	12.5 ± 0.7
	Summer	23.9	25.0	25.6	25.0	23.9	23.1	24.7	24.4	24.0	24.6	24.0	24.4 ± 0.7
	Fall	15.8	15.0	14.2	15.3	12.8	14.8	15.3	15.0	15.8	14.4	15.2	14.9 ± 0.8
Precipitation (mm)	Winter	27	6	21	37	19	23	27	16	20	14	15	20 ± 8
	Spring	96	87	36	16	82	91	90	64	74	101	63	73 ± 26
	Summer	595	321	261	369	323	437	281	241	435	189	811	387 ± 180
	Fall	77	160	75	44	40	123	95	137	30	103	54	85 ± 43
Wind speed (m s^{-1})	Winter	2.3	2.5	2.2	2.2	2.1	2.3	2.5	2.5	2.4	2.1	2.5	2.3 ± 0.2
	Spring	2.4	2.4	2.7	2.5	2.2	2.2	2.7	2.8	2.8	2.7	2.7	2.6 ± 0.2
	Summer	2.2	1.7	1.9	1.3	2.0	1.7	2.3	2.4	2.2	2.4	2.5	2.1 ± 0.4
	Fall	2.0	1.2	1.7	1.4	2.0	1.7	2.1	2.1	2.2	2.3	2.0	1.9 ± 0.3
Wind direction	Winter	NW	WNW	W	W	W	W	WNW	WNW	WNW	WNW	WNW	WNW
	Spring	NWN	W	W	W	W	NE	WNW	WNW	WNW	WSW	W	W
	Summer	ENE	NE	ENE	ENE	ENE	NE	ENE	WNW	NE	ENE	ENE	ENE
	Fall	ENE	Calm	NE	ENE	NE	NE	NE	NE	NE	NE	NE	NE

northerly winds in other cities) (see Table 5 and Table 4S) and air mass back trajectory (not shown, Hybrid Single Particle Lagrangian Integrated Trajectory Model, http://ready.arl.noaa.gov/hysplit-bin/trajtype.pl?runtype=archive). It was found that the contribution of a long-range transport (from China to South Korea) can account for about 20% of SO_2 input [35]. In contrast, analysis of the summer period with dominant ENE, easterly winds in Seoul (and southerly winds in other cities) and air mass back trajectory (from western Pacific Ocean) suggests the occurrence of low CO levels at the U-RS sites in Seoul (and other cities) in compliance with meteorological conditions.

4. Summary and conclusions

In this study, the concentration data of CO measured from 16 urban roadside sites in Korea over an 11-year period were examined to account for a noticeable shift in concentration levels after 2000. A line of evidence collected in this study supports the idea that the changes in CO levels occurred predominantly at roadside stations around Seoul in a tight association with the introduction of the natural gas vehicle supply program in 2000 and improvement of fuel quality. Although the patterns of other cities are not as strong as that of Seoul, the effect of such control efforts has also been effective enough to induce air quality change, at least with respect to carbon monoxide.

An evaluation of CO distribution data at all U-RS stations confirmed the mean reduction rate of 0.093 ppm year^{-1} (or 9.8% year^{-1}) in all 16 U-RS monitoring stations across Korea for the 11 year period. At many stations, the significant reduction in CO levels is recognizable during such a transition period. In addition, the pattern during period II generally resumed temporal stability after noticeable drops in the early 2000 s. Considering the fact that the strong reductions in CO levels were apparent after 2000, the effect of fuel type changes (upgrading) and natural gas engines should have been highly effective strategies in air pollution reduction, especially at the beginning stages. Because of the absence of concurrent measurements on other relevant trace species like CO_2 or VOC, we cannot confirm such an effect from those data sets. However, it is most likely that the comprehensive control efforts should have exerted great influences on CO concentration levels. As a result, it is recommended to develop more effective strategies to implement reduction policies in a highly systematic and consistent manner.

Acknowledgments

This work was supported by a National Research Foundation of Korea (NRF) grant funded by the Ministry of Education, Science and Technology (MEST) (No. 2010-0007876). Corresponding author acknowledges financial support from “The Eco-technopia 21 project” of Korea Ministry of Environment.

Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at doi:10.1016/j.jhazmat.2011.01.099.

References

[1] R.W. Atkinson, B. Barratt, B. Armstrong, H.R. Anderson, S.D. Beevers, I.S. Mudway, D. Green, R.G. Derwent, P. Wilkinson, C. Tonne, F.J. Kelly, The impact of the congestion charging scheme on ambient air pollution concentrations in London, Atmos. Environ. 43 (2009) 5493–5500.
[2] M. Grah, C. Azar, M.I. Williander, J.E. Anderson, S.A. Mueller, T.J. Wallington, Fuel and vehicle technology choices for passenger vehicles in achieving stringent CO_2 targets: connections between transportation and other energy sectors, Environ. Sci. Technol. 43 (2009) 3365–3371.
[3] E. Uherek, T. Halenka, J. Borken-Kleefeld, Y. Balkanski, T. Berntsen, C. Borrego, M. Gauss, P. Hoor, K. Juda-Rezler, J. Lelieveld, D. Melas, K. Rypdal, S. Schmid, Transport impacts on atmosphere and climate: land transport, Atmos. Environ. 44 (2010) 4772–4816.
[4] D. Mage, G. Ozolins, P. Peterson, A. Webster, R. Orthofer, V. Vandeweerd, M. Gwynne, Urban air pollution in megacities of the world, Atmos. Environ. 30 (1996) 681–686.
[5] N.M. Baldasano, E. Valera, P. Jimenez, Air quality data from large cities, Sci. Total Environ. 307 (2003) 141–165.
[6] M.J. Molina, L.T. Molina, Megacities and atmospheric pollution, J. Air Waste Manage. Assoc. 54 (2004) 644–680.
[7] B.R. Gurjar, T.M. Butler, M.G. Lawrence, J. Lelieveld, Evaluation of emissions and air quality in meagcities, Atmos. Environ. 42 (2008) 1593–1606.
[8] P. Hoor, J. Borken-Kleefeld, D. Caro, O. Dessens, O. Endresen, M. Gauss, V. Grewe, D. Hauglustaine, I.S.A. Isaksen, P. Jöckel, J. Lelieveld, G. Myhre, E. Meijer, D. Olivie, M. Prather, C.S. Poberaj, K.P. Shine, J. Staehelin, Q. Tang, J. van Aardenne, P. van Velthoven, R. Sausen, The impact of traffic emissions on atmospheric ozone and OH: results from QUANTIFY, Atmos. Chem. Phys. 9 (2009) 3113–3136.
[9] J.T. Cohen, Diesel vs. compressed natural gas for school buses: a cost-effectiveness evaluation of alternative fuels, Energ. Policy 33 (2005) 1709–1722.
[10] H.T. Nguyen, K.-H. Kim, C.-J. Ma, S.-J. Cho, Long-term study of CO and CH_4 behavior at an urban roadside and urban background locations in Seoul, Korea, Environ. Res. 110 (2010) 396–409.
[11] KMOE (Korean Ministry of Environment), Environmental Statistics Yearbook 2008.
[12] NIER (National Institute of Environmental Research), National air pollutants emission 2007, 2008.
[13] NIER (National Institute of Environmental Research), National air pollutants emission 455 2006, 2007.
[14] NIER (National Institute of Environmental Research), National air pollutants emission 2005, 2006.
[15] NIER (National Institute of Environmental Research), National air pollutants emission 2004, 2005.
[16] NIER (National Institute of Environmental Research), National air pollutants emission 2003, 2004.
[17] NIER (National Institute of Environmental Research), National air pollutants emission 2002, 2003.
[18] NIER (National Institute of Environmental Research), National air pollutants emission 2001, 2002.
[19] NIER (National Institute of Environmental Research), National air pollutants emission 2000, 2001.
[20] NIER (National Institute of Environmental Research), National air pollutants emission 1999, 2000.
[21] EMEP/CORINAIR, EMEP/CORINAIR 2007 Emission Inventory Guidebook, 2007.
[22] NIER (National Institute of Environmental Research), Calculation method of national air 472 pollutant emission, 2005.
[23] K.K. Kang, Environmental policies for fuel switching, Korea Environ. Policy Bull. 2 (2004) 1–19.
[24] W.-K. Jo, J.-H. Park, Characteristics of roadside air pollution in Koran metropolitan city (Daegu) over last 5 to 6 years: Temporal variations, standard exceedances, and dependence on meteorolotical conditions, Chemosphere 59 (2005) 1557–1573.
[25] S.K. Goyal, P. Sidhartha, Present scenario of air quality in Delhi: a case study of CNG implementation, Atmos. Environ. 37 (2003) 5423–5431.
[26] V. Kathuria, Impact of CNG on vehicular pollution in Delhi: a note, Transport. Res. D Transport Environ. 9 (2004) 409–417.
[27] J. Gasca, E. Ortiz, H. Castillo, J.L. Jamies, U. González, The impact of liquefied petroleum gas usage on air quality in Mexico City, Atmos. Environ. 38 (2004) 3517–3527.
[28] A. D'Angiola, L.E. Dawidowski, D.R. Gómez, M. Osses, On-road traffic emissions in a megacity, Atmos. Environ. 44 (2010) 483–493.
[29] B.R. Gurjar, A.S. Nagpure, T.P. Singh, Air quality in megacities, in: C.J. Cleveland (Ed.), Encyclopedia of Earth, Environmental Information Coalition, National Council for Science and the Environment, Washington, DC, 2010.
[30] P.-H. Kuo, P.-C. Ni, A. Keats, B.-J. Tsuang, Y.-Y. Lan, M.-D. Lin, C.-L. Chen, Y.-Y. Tu, L.-F. Chang, K.-H. Chang, Retrospective assessment of air quality management practices in Taiwan, Atmos. Environ. 43 (2009) 3925–3934.
[31] NIER (National Institute of Environmental Research), Analysis of improvement effect of greenhouse gas and air pollutant by enhancement of fuel quality control, 2009.
[32] Korea Energy Economics Institute (KEEI), Monthly energy statistics, 2010.12-2008.01, http://www.keei.re.kr/main.nsf/index.html.
[33] C. Cai, C. Hogrefe, P. Katsafados, G. Kallos, M. Beauharnois, J.J. Schwab, X. Ren, W.H. Brune, X. Zhou, Y. Hed, K.L. Demerjian, Performance evaluation of an air quality forecast modeling system for a summer and winter season – photochemical oxidants and their precursors, Atmos. Environ. 42 (2008) 8585–8599.
[34] J.H. Seinfeld, S.N. Pandis, Atmospheric Chemistry and Physics – From Air Pollution to Climate Change, 2nd ed., John Wiley & Sons, 2006.
[35] S.-U. Park, E.-H. Lee, Long-range transport contribution to dry deposition of acid pollutants in South Korea, Atmos. Environ. 37 (2003) 3967–3980.
[36] G.I. Gorchakow, E.G. Semutnikova, E.V. Zotkin, A.V. Karpov, E.A. Lezina, A.V. Ul'yanenko, Variations in gaseous pollutants in the air basin of Moscow, Izv. Atmos. Ocean. Phys. 42 (2006) 176–190.
[37] C.K. Chan, X. Yao, Air pollution in mega cities in China, Atmos. Environ. 42 (2008) 1–42.