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Progress in the reduction of carbon monoxide levels in major urban areas in Korea

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ABSTRACT

Long-term trends in observed carbon monoxide (CO) concentrations were analyzed in seven major South Korean cities from 1989 to 2013. Temporal trends were evident on seasonal and annual timescales, as were spatial gradients between the cities. As CO levels in the most polluted cities decreased significantly until the early 2000s, the data were arbitrarily divided into two time periods (I: 1989–2000 and II: 2001–2013) for analysis. The mean CO concentration of period II was about 50% lower than that of period I. Long-term trends of annual mean CO concentrations, examined using the Mann–Kendall (MK) method, confirm a consistent reduction in CO levels from 1989 to 2000 (period I). The abrupt reduction in CO levels was attributed to a combination of technological improvements and government administrative/regulatory initiatives (e.g., emission mitigation strategies and a gradual shift in the fuel/energy consumption mix away from coal and oil to natural gas and nuclear power).

1. Introduction

Carbon monoxide (CO) is predominantly the result of incomplete combustion, and has both natural and anthropogenic sources (e.g., forest fires and internal combustion engine emissions). The combination of its physical characteristics (colorless, odorless, and tasteless) and severe health risks has made CO a notorious poisonous gas in confined spaces (Lindell and Weaver, 2009; Choi et al., 2014); it is also considered an important ambient (outdoor) air pollutant, affecting the oxidation capacity of the atmosphere with a relatively long lifetime. It should be noted, however, that the ambient air CO concentrations reported herein are well below the toxic levels to humans or animals (Goldsmith and Landaw, 1968; Weaver, 2009; Lindell and Weaver, 2009).

Vehicle emissions not only cause primary atmospheric pollution (e.g., CO, HC, and NO_x) but also result in the production of secondary pollutants like O_3 and peroxyacetyl nitrate (PAN) through photochemical reactions. CO emissions from gasoline fueled vehicles are particularly high when engines are idling, since the efficiency of catalytic converters decreases to less than 10% when engine exhaust temperatures are less than optimal (200–300 °C) (http://bpm.kemco.or.kr/transport/dr/dr_02.asp). Consequently, CO is a dominant constituent of congested “traffic jam” emissions (Stedman, 1989; Chan et al., 2002).

The Republic of Korea (RoK) has a population of 49 million, a density of 489 persons km^{-2}, with 83.8% living in urban areas, and 64% of the land area is forested (2011–2013 UNdata, URL: https://data.un.org/). The RoK's population (in millions) increased from 31.4 (1970), 40.5 (1985), 46.0 (2000), to 49.8 (2015 (estimation)); it is projected to decrease from a maximum of 52.4 after 2035 (http://esa.un.org/wpp/unpp/p2k0data.asp). Since ca. 1980, the RoK has undergone very rapid economic development as reflected in various energy indicators of economic activity. For example, (a) CO_2 emissions (Mt CO_2): 132 (1980)–657 (2012), (b) natural gas consumption (billion cubic feet): 0 (1985)–1877 (2012), (c) oil consumption (barrels): 552,000 (1985)–2,340,000 (2012), and (d) coal consumption (million short tons): consumption varied from 30.8 (1980)–53.9 (1986), dipped to 43.1 (1992) and steadily increased since to 137.6 (2012). In 2012, renewable energy and nuclear power represent 1% and 12%, respectively, of the energy mix (U.S. Energy Information Administration). The number of registered vehicles

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rose from ~2,000,000 (1988) to ~20,000,000 (2014) (http://www.tradingeconomics.com/south-korea/car-registrations). In spite of the huge increase in fossil fuel consumption and number of registered vehicles, RoK nationwide CO emissions remained fairly constant at 820 ± 68 kt CO per year for the period 1999–2012; in contrast, nationwide CO emissions decreased by a factor of ~2 from 1991 kt CO/y (1990) to 977 kt CO/y (1998) (National Institute of Environmental Research (2012)).

In light of the worldwide recognition of CO's environmental significance, the Korean Ministry of the Environment (KMOE) established environmental standards for CO as well as a suite of other criteria pollutants. A network of monitoring stations has subsequently been commissioned comparable to those of developed (OECD) countries including Japan, the USA, the Netherlands, and Germany. Since 1983, the South Korean domestic guideline values for ambient CO levels have been set at 25 and 9 ppm for 1- and 8-h averages, respectively (White paper on the environment, KMOE, 2013). Furthermore, regulatory guidelines for five pollutants (including CO) were implemented to improve indoor air quality (IAQ) in multi-use facilities (e.g., subway stations and underground parking lots) and new apartment buildings. According to the revised IAQ law of December 19, 2011, the standard for indoor CO concentrations is 10 ppm (in facilities such as subway stations, libraries, medical institutions, etc.) and 25 ppm (in parking lots).

Because of the growing interest in air quality management for major pollutants, CO has been extensively studied worldwide (e.g., WHO website). In our previous study (Kim and Shon, 2011), we analyzed CO concentrations from seven major South Korean cities between 1998 and 2008 in order to investigate the spatiotemporal characteristics. In this work, we expand upon our previous study by investigating a substantially extended period (1989–2013), CO correlations with selected priority pollutants (i.e., SO_2, NO_2, O_3, and TSP), contribution from briquette CO emissions, and reference background CO levels. Consistent with our previous study, the entire 25-year dataset was divided into two time periods (I: 1989–2000, and II: 2001–2013), centered on the early 2000s, the end of the large reduction in observed CO levels. The spatiotemporal characteristics of CO concentrations across South Korea are statistically analyzed and compared between these time periods. It is intended that the results of our analysis will help to develop systematic strategies and policies leading to improved standards to reduce CO emissions. In this way, the concentration of CO in ambient air can be further reduced to a more manageable level.

2. Methodology

2.1. Site characteristics of the study areas

To investigate the spatial distribution and temporal trends of CO in South Korea, its concentration was monitored in seven major cities (Seoul, SL; Busan, BS; Daegu, DG; Incheon, IC; Daejeon, DJ; Gwangju, GJ; and Ulsan, UL; Fig. 1) over more than two decades (1989–2013) and split into an early (1989–2000) and a late (2001–2013) study period for simplicity. All seven cities have populations over 1.5 millions and have designated as special areas by the RoK government for air pollution monitoring and control (http://eng.me.go.kr/eng/web/main.do, accessed August 2015). Table 1S shows the number of air pollution monitoring stations operated in each of the seven cities throughout the study period. It should be noted that the total number of monitoring stations steadily increased from 21 in 1989 to 98 in 2013.

South Korea is a peninsula on the north-eastern edge of the Eurasian landmass, with a temperate climate consisting of four very distinct seasons that vary from a cold, dry winter (−6–3 °C, January), to a hot, humid summer (23–26 °C, July/August). The Köppen climate classification codes for South Korea are Dwa, Cfa, and Cwa; between which the cities are divided as: Seoul and Incheon (Dwa); Daegu, Daejeon, and Gwangju (Cwa); and Busan and Ulsan (Cfa). The prevailing wind patterns are generally south-easterly in summer and north-westerly in winter (Zahorowski et al., 2005; see also website: http://www.asianinfo.org/asianinfo/korea/geo/climate_and_weather.htm).

Seoul is the capital of South Korea, and the largest metropolitan area. In 2004, the population of SL was 10,036,241 and incurred no significant population growth since (National Statistical Office, 2014). Busan, located on the south-eastern-most tip of the Korean peninsula, is the second largest city. BS is the country's main port for international trade (world's fifth busiest port) with a population exceeding 4 million. DG, Korea's third most populous city, is located in south-eastern part of Korea near the Nakdong River. IC is a port city, designated as Korea's first free economic zone in 2003. Since then, a large number of both national and international companies have increasingly invested in the Incheon Free Economic Zone (IFEZ). Located in the center of South Korea, DJ is a transportation hub, which had a population of over 1.5 million in 2010. DJ is considered to be a science and technology center with the Daedeok Research and Development Special Zone (28 state-run research centers as well as 79 private enterprise research institutes). UL is a highly industrialized city located in the south-eastern part of the Korean Peninsula. UL is the industrial powerhouse of South Korea with two enormous industrial complexes within its city limits, namely, the Ulsan petrochemical complex and the Ulsan Mipo Industrial Complex (Lee et al., 1999; Nguyen and Kim, 2006).

2.2. Carbon monoxide measurements

Hourly measurements of CO concentration were made at each monitoring station. These data were subsequently converted into monthly means for each city by the Ministry of Environment (http://airemiss.nier.go.kr/main.jsp). CO concentrations were measured using the non-dispersive infrared (NDIR) method (ZRF, Fuji Electric Co., Ltd., Japan), which involves sensing infra-red absorbance changes using a selective detector (NIER, 2008). From the measured absorbance of these gaseous contaminants, their concentrations were calculated based on the Lambert–Beer law principle. The ZrF_4 (ZRF) Infrared Gas Analyzer is generally known to maintain high accuracy, high sensitivity, and stability over time. The practical CO concentration range of this instrument is 0.1–100 ppm at a sampling flow rate of 0.5 ± 0.25 L/min. The lower detection limit of CO is 0.05 ppm with full scale accuracy of ±0.5%.

3. Results and discussion

To assess the spatiotemporal characteristics of CO, we relied on monthly mean concentrations collected from seven major South Korean cities from 1989 to 2013 as the basic criteria used in the statistical analysis. A primary intention of this analysis is to characterize the long-term (decadal) and short-term (seasonal) temporal CO trends, with particular emphasis on measurements after 2000, when the concentrations of CO became much lower than during the earlier period of this study (Fig. 2).

3.1. Regional CO concentration patterns

Table 1 provides a statistical summary of annual mean CO concentrations (ppb) in seven major South Korean cities based on monthly mean data from the individual stations. The average CO concentration across all seven cities in 1989 was 2113 ppb, which steadily decreased to 505 ppb by 2013 (a decrease of about 76%). The seven-city averaged CO concentration in 2013 (505 ppb) was

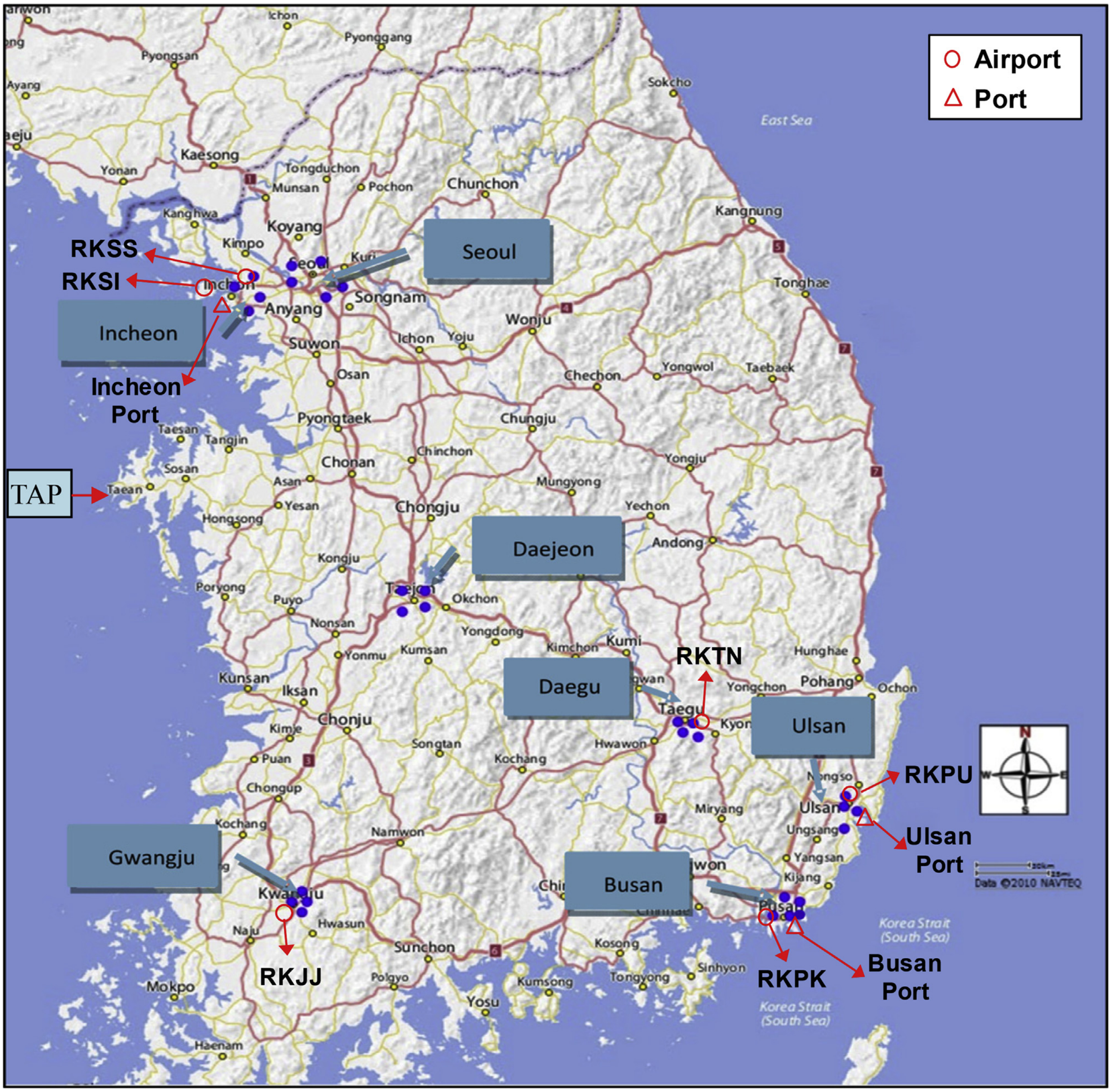

Fig. 1. Map showing the seven major cities sites and location of the Tae-Ahn peninsula (TAP) in South Korea (based on NAVTEQ map).

nonetheless considerably higher than the regional background level for Korea (241 ± 47 ppb: range 136–378 ppb) recorded between 1991 and 2013 at the rural coastal site on the Tae-Ahn Peninsula (TAP) (Fig. 1, Figs. 1S, and 2S). The TAP site (~120 km SW of Seoul, 36.7376° N, 126.1328° E) is part of NOAA's Earth System Research Laboratory – Global Monitoring Division network. The largest observed decrease in CO levels from 1989 to ca. 2004 (Fig. 2) may in-part reflect the effect of the implementation of more stringent air quality and emissions standards for CO since 1983 (http://eng.me.go.kr/eng/web/main.do, accessed August 2015).

The summary of CO data in Table 1 allowed the comparison of variations among three periods: period I (1989–2000), period II (2001–2013), and the entire dataset (1989–2013). For the entire 1989–2013 period, among the 7 cities studied, Seoul recorded the highest annual mean value of CO (1112 ppb) with lower concentrations in Daegu (935 ppb) and Incheon (933 ppb). These values are all higher than the 7-city average (919 ppb). The higher value in Seoul is likely attributable to the large number of gasoline and diesel vehicles, as it has the maximum number of registered cars (per km^2) in South Korea (KMOE, 2006). Seoul is located in the Han River valley, flanked by low mountains, potentially leading to air stagnation (Lee et al., 2008; Park et al., 2004).

3.2. Seasonal patterns of CO concentration in relation to local sources

Analysis of the CO data clearly indicated a greater temporal than spatial variability among all cities for all study periods (Table 1). Based on data from all cities, the mean annual CO was 1310 and 583 ppb, for periods I and II, respectively (a decrease of 56%). Considering the individual stations within each city, mean annual CO concentrations showed pronounced maxima of 2100–5100 ppb

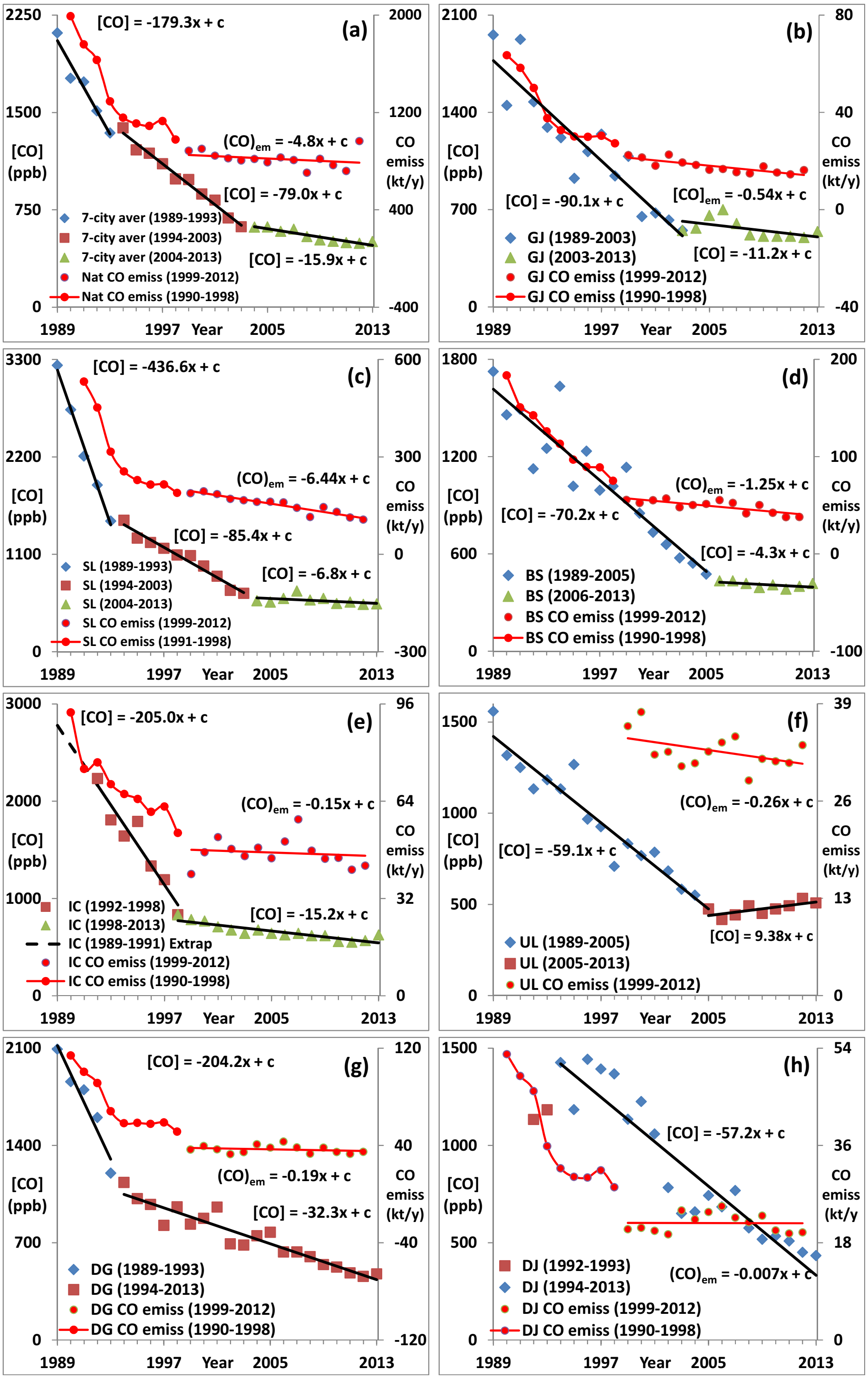

Fig. 2. Trends in measured annual CO concentrations (primary y axis) and CO emissions (secondary y axis) nationally and at seven major cities in South Korea. The CO emissions data are plotted as two separate periods (1990–1998) and (1999–2012) with no merging.

and 1000–2000 ppb for periods I and II, respectively (Table 2S).

Seasonal CO cycles (Fig. 3S – Seoul) indicate that its mean concentrations are the highest in winter for periods I and II in all seven cities. This observed pattern of CO is fairly comparable to that of SO_2 in some senses (Fig. 4S). In general, the temporal cycle of CO, if evaluated in terms of its seasonal mean values from all seven cities, was characterized by the following descending order: winter > fall > spring > summer. These seasonal results from all seven cities can also be compared between the two periods. The summer-term results during period I and II (925 and 483 ppb) were as low as 50% of their winter counterparts (1796 and 810 ppb). If this type of comparison is extended further to other seasons, the respective results were quite comparable between spring (1331 and 602 ppb) and fall (1361 and 626 ppb). Consumption of fossil fuels (e.g., natural gas) in winter is approximately a factor of 6.5 higher than in summer, due to the winter maximum in domestic heating

Table 1

Statistical summary of annual mean CO concentrations (ppb) in seven major South Korean cities[a] (using monthly mean data).

	Year	CO(ppb)							Average
		SL	BS	DG	IC	GJ	DJ	UL	
A. Period I (1989–2000)	1989	3233	1725	2092	–[b]	1958	–	1558	2113
	1990	2733	1458	1858	–	1450	–	1317	1763
	1991	2208	1492	1800	–	1925	–	1250	1735
	1992	1883	1125	1600	2233	1475	1133	1133	1512
	1993	1475	1250	1200	1808	1292	1183	1183	1342
	1994	1483	1633	1133	1642	1217	1425	1133	1381
	1995	1283	1017	1017	1792	925	1183	1267	1212
	1996	1233	1233	975	1333	1117	1442	967	1186
	1997	1167	992	825	1192	1242	1392	925	1105
	1998	1092	1017	958	833	942	1367	708	988
	1999	1083	1133	833	783	1083	1133	833	983
	2000	967	850	875	767	650	1225	767	871
	Mean	1653	1244	1264	1376	1273	1276	1087	1310
	SD	920	488	654	629	713	355	344	586
	N	144	144	144	108	144	108	144	134
	Range (Median)	600–5000 (1400)	500–2600 (1100)	500–5100 (1200)	500–3400 (1200)	400–3800 (1050)	600–2200 (1300)	500–2100 (1000)	514–3457 (1178)
B. Period II (2001–2013)	2001	850	733	957	708	675	1058	786	824
	2002	692	658	692	675	625	783	683	687
	2003	658	575	683	642	550	650	583	620
	2004	575	542	750	675	567	658	550	617
	2005	558	475	775	642	658	742	475	618
	2006	600	433	633	625	700	683	417	585
	2007	683	433	633	642	600	767	442	600
	2008	583	417	600	617	517	575	492	543
	2009	600	392	542	617	508	517	450	518
	2010	542	408	525	558	508	533	475	507
	2011	558	383	483	550	508	508	492	498
	2012	533	400	458	567	500	450	533	492
	2013	533	417	475	625	542	433	508	505
	Mean	613	482	621	626	574	643	521	583
	SD	184	135	207	151	169	258	142	178
	N	156	156	151	156	156	156	151	155
	Range (Median)	400–1200 (600)	300–1000 (500)	0–1300 (600)	400–1000 (600)	100–1100 (600)	400–2000 (600)	200–1000 (500)	243–1229(571)
C. All period (1989–2013)	Mean	1112	848	935	933	909	902	797	919
	SD	833	518	578	557	617	433	385	560
	N	300	300	295	264	264	264	295	288
	Range (Median)	400–5000 (800)	300–2600 (700)	0–5100 (700)	400–3400 (700)	400–3400 (700)	300–2200 (800)	200–2100 (700)	243–3457(729)
Ratio of mean (Period I/II)		2.70	2.58	2.04	2.20	2.22	1.98	2.09	2.26

[a] Seoul = SL, Busan = BS, Daegu = DG, Incheon = IC, Gwanju = GJ, Daejeon = DJ, Ulsan = UL.

[b] Not available.

requirements (Korea Development Institute, http://www.kdi.re.kr/kdi_eng/main/main.jsp). Another factor is an increase in frequency of atmospheric stagnation events in winter due to persistent anti-cyclonic conditions (Lee et al., 2008; Park et al., 2004).

According to satellite-borne measurements, during the boreal spring, the Eastern China region has the highest average CO level of 300 ppb among the world regions (http://neo.sci.gsfc.nasa.gov/dataset_index.php#atmosphere). On the other hand, the Pacific Ocean well off the coast of Japan has a boreal summer CO level of ~100 ppb. Therefore, the seasonal change in prevailing wind direction is responsible for a difference of about 200 ppb supported in part by measurements at the TAP observatory (Figs. 1S and 2S). Likewise, there is a notable difference in CO levels between winter and summer in Period II, an average decrease of ~325 ppb observed for all 7 cities in this study and about ~125 ppb can be ascribed to seasonality of natural gas usage. During Period I, spring/summer differences were generally much larger and more variable (e.g., from ~575 ppb (SL) to ~123 ppb (UL)). In 2013, SL had a mean annual average CO level of 533 ppb. If we assume that averaged annual CO concentrations at TAP represent a background value (in Seoul), it can be inferred that average annual CO concentrations of Seoul in 2013 can be apportioned as ~240 ppb (45%) from ambient background and ~285 ppb (55%) from anthropogenic sources. As the natural Asian continental background CO level in Western China, Mongolia, and Kazakhstan is about 100 ppb (http://www.esrl.noaa.gov/gmd/ccgg/flask.php), this predicts the lowest CO level possible in South Korea, if all anthropogenic global CO sources are eliminated, globally. Unless CO pollution emissions in China are curtailed, the CO level in SL can only be reduced to ~240 ppb even if all CO anthropogenic sources in SL are eliminated.

Similar seasonal cycles of CO concentration have been extensively noted and reported worldwide: e.g., in the USA, Japan, Hong Kong, China (e.g., Beijing), India, Canada, and South Korea (NOAA GMN (http://www.esrl.noaa.gov/gmd/ccgg/flask.php)). In addition, the same pronounced seasonal trends have been observed by NOAA's trace gas global monitoring network of stations located at sites remote from large urban areas (http://www.esrl.noaa.gov/gmd/ccgg/flask.php). Monthly CO levels recorded at 4 NOAA monitoring sites from ca. 1990 to 2013, i.e., Mauna Loa Observatory (MLO), HI, USA; Tae-ahn Peninsula (TAP), South Korea; Cape Grim (CGO), Tasmania, Australia; and South Pole (SPO), Antarctica are shown in Fig. 1S. The yearly mean CO level at each of the 4 sites showed no significant temporal change. The average CO levels for all years are 241 ± 47 ppb (TAP), 93 ± 19 ppb (MLO), 52 ± 8 ppb

(CGO), and 49 ± 9 ppb (SPO) unlike the much higher levels observed for all 7 South Korean cities in this study.

In Fort Meade (Maryland, USA), the seasonal CO cycle was characterized by high concentrations in winter and low concentrations in summer (Chen et al., 2001), with seasonal mean CO concentrations descending as: winter > fall > spring > summer. In Oki, Japan, seasonally averaged CO concentrations revealed a pronounced spring maximum (208 ppb) in the continental background air mass compared to the summer minimum (120 ppb). By comparison, seasonal average CO concentrations of the regionally polluted continental air showed a spring maximum of 271 ppb and summer-autumn minimum of 180 ppb (Pochanart et al., 1999). In Hong-Kong, a very distinct seasonal variation in CO levels was observed, with a low summer minimum (116 ppb) and high winter maximum (489 ppb) (Lam et al., 2001). In Beijing, CO levels were higher in winter compared to spring, summer, and fall (Chan and Yao, 2008; Wang et al., 2010). Similarly, in New Delhi, India, and Toronto, Canada, the concentration of CO showed a summer minimum and a winter maximum (Aneja et al., 2001; Burnett et al., 1998; CPCB).

3.3. *Long-term (inter annual) trends of CO*

To learn more about the long-term trend of CO, we conducted a Mann-Kendall (MK) test using the annual mean CO concentrations over the whole study period for each city. Plots of the long term trends in mean annual CO concentrations for all cities are shown in Fig. 2. Higher overall CO concentrations were seen in SL, DG, GJ, IC (Fig. 2) compared to BS, DJ, US (Fig. 2). For all seven cities, CO concentrations decreased from 1989 to 2013. Annual mean values were significantly different between period I (1310 ppb) and period II (583 ppb). However, while the annual mean values decreased considerably every year throughout period I, they were similar, or decreased at a reduced rate, throughout period II.

Linear trend analysis of the CO concentration data (Fig. 2) showed three distinct decay periods for Seoul and the 7-city average; the corresponding decay rates were: −437 and −179 (1989–1993), −85 and −79 (1994–2003), and −7 and −16 (2004–2010) ppb/year, respectively. The sub-period 1989–1993 had the largest annual decay rate, e.g., Seoul (−437 ppb/year) and whereas the 2003–2010 sub-period showed the smallest decay rate, e.g., Seoul (−7 ppb/year). On the other hand, the (2000–2012) decadal satellite observations on the monthly column CO densities in the Northern Hemisphere (0–60°N) and over East China showed only insignificant decay over time (Worden et al. 2013); this is in excellent agreement with ground-based measurements at TAP (see Fig. 3S).

Most other cities (IC, DG, GJ, BS, and UL) showed two distinct decline periods (an initially fast decline followed by slower decline) as shown in Fig. 2c–e, g. The city, DJ, (Fig. 2h) showed only a fast CO decline (63 ppb/year) for the 1994–2010 period. Interestingly, UL, for unknown reasons, showed a small increase of 9.4 ppb/y for the 2005–2013 period (Fig. 2f). The length of the two decline periods varied from city to city. For the fastest decline period, the CO decline rates (ppb/year) were 437 (SL), 205 (IC), 204 (DG), 179 (7-city average), 90 (GJ), 70 (BS), and 59 (UL). For the slowest decline period, the decline rates (ppb/year) ranged from 0 to 10 (BS, SL) and 10–20 (GJ, IC, 7-city average). Two cities, DG and DJ continued to show moderately fast declines of −32 and −57 ppb/y, respectively up to 2013.

Also shown in Fig. 2 (on the secondary y axis, kt CO/year) are the corresponding annual CO emission rates nationally and for each city split into two separated periods of (1990–1998) and (1999–2012). The UL CO emission data for the (1990–1998) period are incomplete and hence not shown in Fig. 2f. On the national level, CO emissions for the 1999–2012 period were fairly constant at 820 ± 68 kt CO/year (Fig. 2a) even though the 7-city average CO levels decreased significantly (−79 and −16 ppb/year). The decline in CO emissions (kt CO/year/year) ranged from ~0 (nationally, IC, DG) to 6.4 (SL). During this period, Seoul (and other cities) showed only a weak correlation between CO emissions and CO levels for the 1999–2012 period (Fig. 2). In summary, the decline in the observed CO levels after ca. 2000 is due to unknown factors only weakly related with decreasing CO emission rates. On the other hand for the 1990–1998 period, there are reasonable correlations between CO emissions and CO levels for six cities (UL not included as discussed) by visible inspection as seen in Fig. 2.

Such decreases in CO concentrations can be attributed to the implementation of regulatory action and enforcement (e.g., strengthened atmosphere environmental standards in 1995) (White paper on the environment, KMOE, 2013). Also, after 1999, CO emissions from road transport (per vehicle) have continuously decreased, which has helped to maintain CO concentrations below the KMOE environmental standards. South Korean CO emissions from all sources remained relatively stable at 820 ± 68 kt for the 1999–2012 period (National Institute of Environmental Research) despite road vehicle numbers increasing by 60.5% (National Statistical Office, 2013; The Korea Transport Institute, 2013) and gross domestic product (GDP) increasing by ~200% since 1990 (National Statistical Country index, National Statistical Office, 2010). In comparison, road vehicle numbers increased for the period 2000 to 2010 by 12% in England and 6.4% in the U.S. (NEI). In Moscow (Russia), the CO concentration decreased slightly over a 20-year period (1986–2005) despite a quadrupling in the number of road vehicles (Gorchakow et al., 2006; Rakitin et al., 2011; Yurganov et al., 2011). In England (a low-growth mature OECD economy), total annual CO emissions decreased ~3-fold from 5126 kt in 1999 to 1508 kt in 2012 (UK NAEI (http://naei.defra.gov.uk/reports/reports?report_id=801), accessed: Dec. 2014). In the U.S., total annual CO emissions decreased ~2-fold from 114,541 kt in 1999 to 73,433 kt in 2013 (accessed: Feb. 2014, http://www.epa.gov/ttn/chief/trends/index.html). In the EU28, annual CO emissions decreased from 66,000 kt in 1990 to 2200 kt in 2012. Overall, in most OECD countries, annual CO emissions decreased between 2- and 3-fold from 1990 to 2012 (http://www.eea.europa.eu/publications/lrtap-2014, accessed January 2015).

The use of coal briquettes (Yeontan) for heating purposes (mainly by low-income families - (Park and Kwon, 2011)) has decreased dramatically from 18.8 Mt in 1990 to 1.2 Mt in ca. 2002 and increased slightly to 1.9 Mt in 2012; in 2012, the RoK consumed 81% of global coal briquette production (http://unstats.un.org/unsd/energy/balance/concepts.htm, accessed August 2015). Since

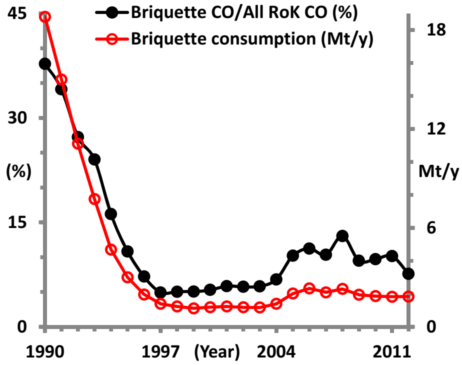

Fig. 3. Percentage contribution of coal briquette CO emissions to total RoK CO emissions inventory and briquette consumption (Mt/y) from 1990 to 2012.

1985, the use of solid fuels for heating purposes (e.g., briquettes) has been increasingly banned; from 1999, it has been banned in 20 regions including Seoul and the six major metropolitan cities of this study (http://eng.me.go.kr/eng/web/main.do, accessed August 2015). On a historical note, many low-income Koreans were killed by CO poisoning (commonly known as “briquette gas”) in the period 1970–1990 when wood was replaced by coal briquettes as fuel for the traditional flue gas “ondol” underfloor heating system; more recently, underfloor heating is now commonly done by a hot water boiler system.

For estimating CO emissions due to the consumption of briquettes, we have used an averaged emission factor of 40 g CO per kg briquette (Shen and Xue, 2014). Coal briquette CO emission factors ranged from 18 to 68 g CO per kg (Zhang et al., 1999). Fig. 3 depicts our estimated contribution of coal briquette combustion CO emissions in relation to the total national RoK CO emissions and consumption for the period 1990–2012. As it can be seen, briquette CO contribution has dramatically decreased from 38% (1990) to a minimum of 5% (1997–2000), while there was a slight increase to about 6–13% (1998–2012). In summary, the phasing out of coal briquettes has substantially reduced overall CO emissions for the period 1990 to 1997 (e.g., see Fig. 2c for Seoul).

3.4. Factors controlling the pollution of CO in the study area

To understand the spatial distribution characteristics of airborne CO, a Pearson correlation analysis was carried out using the annual mean data from the seven cities (Fig. 5S). The strongest correlations with Seoul were for Daegu ($r = 0.9441$), Incheon ($r = 0.9152$), Gwangju ($r = 0.8647$), and Ulsan ($r = 0.8401$). Spatial correlation analysis is an important tool that can direct policymakers to areas where the emission policies have not had the desired effect.

In order to study the relationship between CO and common air pollutants (i.e., SO_2, NO_2, O_3, TSP, and PM_{10}), a correlation analysis was conducted using monthly pollutant data collected in the seven major cities from 1989 to 2013 (Table 3S). The monthly averaged CO and SO_2 levels are in excellent phase with each other as shown in Figs. 2S and 4S, respectively. This implies that both CO and SO_2 have a common source such as coal-fueled power stations (Ray and Kim, 2014). In the Seoul Metropolitan region and Incheon, the increasing use of clean fuels (e.g., LNG, LPG, and low-sulfur diesel) and phasing out of solid fuels has been mandated (http://eng.me.go.kr/eng/web/main.do, accessed August 2015); Consequently, both CO and SO_2 emissions have decreased over time (e.g., see Fig. 4S). If the study period is divided into two periods (period I, period II), minor differences were observed. Generally, CO exhibited its strongest correlations with SO_2, total suspended particles (TSP), and O_3 as shown in Fig. 6S for all seven cities. For example, using an arbitrary correlation criteria of $r > 0.5$ ($P = 0.01$), CO concentration exhibited strong correlations with SO_2 ($r = 0.701$) and TSP ($r = 0.536$). TSP, CO, and SO_2 are generally produced from industrial processes and fossil fuel combustion; CO and SO_2 hence showed significant correlations (Marković et al., 2008). In contrast, O_3 concentrations exhibited a modest inverse correlation for most cities (Fig. 6S). Consequently, CO and SO_2 levels were higher during winter than summer, while O_3 levels were higher during summer and spring than winter (Barrero et al., 2006).

Long-term temporal trends in CO concentration within the same time period as our study (1998–2008) can also be found at two sites abroad: Marylebone Road, London, UK (kerbside monitoring on a busy city center road) and Hohepeissenberg, Germany (high-altitude mountain site). The results of these two sites can be meaningfully interpreted. At Marylebone, annual mean CO concentration decreased at a rate of 12% per year from ~2000 ppb in early 1998 to 530 ppb in 2008, whereas that of VOCs (0.1–30 ppb in 1998) decreased between 3 and 26% per year (Schneidemesser and Monks, 2010). At the high-altitude Hohepeissenberg site, well suited to regional background observations, CO concentrations between 1998 and 2008 were ~10 times lower (~200 ppb) with no clear interannual trend (Schneidemesser and Monks, 2010); This result is very similar to that observed at the Korean background station of TAP, as shown in Fig. 3S.

The various government policies to reduce airborne CO levels appear to exert a major knock-on effect to decrease other air pollutants, as shown by their positive correlations with CO. In 2011, on-road vehicles contributed 64.6% to all RoK CO emissions. In the meantime, the Seoul Metropolitan region accounted for 41.1% of the total national on-road vehicle emissions (http://eng.me.go.kr/eng/web/main.do, accessed August 2015), although it represented ~20% of the RoK's population. The megacity capital area has numerous CO emission sources including all types of road transport vehicles. Therefore, through various government policies and reduction in car tailpipe emissions (e.g., incentives to replace decrepit on-road vehicles with more eco-friendlier vehicles), it was possible to substantially reduce CO emission to the atmosphere (Kim and Shon, 2011).

3.5. The status of CO pollution in South Korea

According to the “Notification on using clean fuel” issued by the South Korean Ministry of Environment in 1988, the use of the alternative fuel, liquefied natural gas (LNG) was mandated for large business furnaces boilers in Seoul to cause the phasing out of coal briquettes. The use of cleaner energy sources by businesses, apartment buildings, and power plants has increased since 2001 (Nguyen et al., 2010a,b). After the 1988 Seoul Olympics, the installation of catalytic converters on new automobiles and the phasing-out of leaded gasoline were implemented by new Ministry of Environment regulations on new cars and unleaded gasoline. At the same time, stricter monitoring of air pollution was also introduced, and non-compliant (decrepit) on-road vehicles were removed from the fleet. Concurrently, pollution management of automobile tailpipe emissions and tighter automobile production methods have all resulted in reduced tailpipe emissions of CO. However, by the late 1990s, traffic flow management became increasingly more emphasized (White paper on the environment, 2002). Additional regulations on car gaseous emissions were implemented as follows: (1) source measures - developing engines with lower emissions and higher thermal efficiency, (2) traffic management - traffic control and restrictions, and (3) nationwide public relations program promoting - cycling, walking, carpooling and public transport (Nguyen et al., 2010a,b). After completing the conversion of city buses to compressed natural gas (CNG) (1991–1997) and the installation of a comprehensive natural gas supply grid in 1998, in large cities city-wide CNG bus route networks were then established. Accordingly, due to revisions of the Clean Air Conservation Act in April 1999, the government set the legal basis for the conversion/replacement of diesel fueled city buses to CNG and ensured the availability of CNG buses. As a consequence, the various government policies on pollution have all contributed to the decrease in CO concentration after 2000 (e.g., stricter enforcement measures, more stringent exhaust emissions standards, and air quality standards) (Shon and Kim, 2011).

Based on the location of South Korea, the air pollution status and industrial activities in Eastern China (if current trends persist) will have a significantly increasing relative impact on the distribution of CO levels in South Korea since stronger CO emission mitigation policies are being implemented in South Korea (Choi and Chang, 2006; Kim and Chung, 2006). Biomass combustion of either artificial or natural materials emits large quantities of pollutants. As biomass and fossil fuel combustion has been steadily increasing in China (and SE Asia in general), it poses a significant threat to the air quality environment of South Korea by increasing the ambient background CO concentrations from less than 100–~300 ppb (http://neo.sci.gsfc.nasa.gov/view.php?datasetId=MOP_CO_M; accessed January 2015). Therefore, much attention is now being paid to the significance of regional pollution (e.g., biomass combustion) (Choi and Chang, 2006). Choi and Chang (2006) used 3-day HYSPLIT back-trajectories to assess the impact of biomass combustion in central and east Siberia (forest fires only) in April 2000 on CO concentrations over the East Japan Sea. They concluded that Siberian forest fires increased CO by ~30 ppb compared to the 11 days when the air masses were non-Siberian in origin. At Happo, Japan, a similarly observed effect of Siberian forest fire on the change in [CO] of 15–47 ppb (April to September 1998) was observed; air masses originating from the Pacific Ocean (June–September, 1998) had a CO concentration of ~110 ppb (Kato et al., 2002).

A comparison of CO levels in various world regions is presented in Table 2S together with our results for periods I and II. The concentrations of CO in Japan, London, and Moscow are similar to the annual mean in Korea during 1998–1999. In contrast, certain regions of Europe and North America differ greatly compared to South Korea. Taiwan and Japan show similar concentrations as Korea. The CO levels in Great Britain are much lower than that of Korea, whereas CO concentrations in North America (USA and Canada) are higher than that of Korea, although the environmental criterion was not exceeded. The annual mean CO levels are higher by factor of 2 in urban areas than those in coastal areas in Taiwan (Lin et al., 2008). The urban CO levels in Taiwan (Chungmin) are higher than South Korea but the coastal areas in Taiwan (Erhlin) are lower than South Korean urban areas. Therefore, CO levels in South Korea are similar to the values of other East Asian countries, as South Korea is greatly affected by China due to geography.

According to the National Ambient Air Quality Standards (NAAQS standards) of the US Environmental Protection Agency (EPA), the standard (time weighted) CO exposure limit is set at 9 ppm for an 8-h period and 35 ppm for a 1-h period. Also, atmosphere environmental CO standards in various countries are: 10 ppm for a 24-h period, 20 ppm for an 8-h period (Japan); 8.6 ppm for an 8-h period (European Union), 9 ppm for an 8-h period, 25 ppm for a 1-h period (Hong Kong), and 3.2 ppm for an 24-h period, 8 ppm for a 1-h period (China). The corresponding atmosphere environmental CO standards in South Korea are 9 ppm for an 8-h period, 25 ppm for a 1-h period. Thus, the South Korean standards are comparable to the U.S, Japan, European Union, Hong Kong and China. In South Korea, the mean annual CO levels (e.g., 0.41–0.56 ppm in 2010) reported herein since 1990 have all been much less than the environment standard (8-h time weighted, 9 ppm).

4. Conclusions

The temporal and spatial variability of CO concentrations were statistically analyzed using observations from seven major South Korean cities during the period 1989–2013. Although each city showed regional concentration differences, the seasonal or periodical change was more notable. On sub-yearly timescales, winter CO concentrations were found to be approximately two times higher than summer values. This observation was influenced by various factors, e.g., a major cause is ascribed to natural gas domestic heating in winter. Analysis of the long term trends for two time periods of I (1989–2000) and II (2001–2013) indicated that CO levels were 2 times higher in the former than the latter.

Also, the concentration of CO between cities during the 1989–2013 period indicates significant decrease after 2000. This variation may be affected by administrative control efforts such as fuel conversion policy and strengthening of atmosphere environmental standards. Through strict government regulations and enforcement, CO levels have greatly declined. Therefore, we need to continue with diverse policies to consistently decrease CO concentration, e.g., the levels achieved in the EU.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.envpol.2015.09.008.

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