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

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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

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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).

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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) CO₂ emissions (Mt CO₂): 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







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 \pm 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 1and 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., SO₂, NO₂, O₃, 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 ($-6-3 \ ^{\circ}C$, January), to a hot, humid summer ($23-26 \ ^{\circ}C$, 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 southeasterly 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 ZrF4 (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 \pm 0.25 L/min. The lower detection limit of CO is 0.05 ppm with full scale accuracy of \pm 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

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