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Long-term trend of NO_2 in major urban areas of Korea and possible consequences for health

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HIGHLIGHTS

- Long-term variations of NO_2 are investigated using the data from seven major urban areas in Korea.
- The pollution status of NO_2 is explored in association with dynamic changes in environmental conditions.
- The observed reductions after 2000 reflect the direct effects of implementation of administrative efforts.

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ABSTRACT

Long-term trend of the atmospheric NO_2 was analyzed using ambient monitoring data collected from seven major cities in Korea over two decades (1989–2010). In light of the notable environmental policies initiated since June 2000, these NO_2 data were also evaluated after dividing the entire study period into period I (1989–1999) and period II (2000–2010). Accordingly, the mean concentrations of NO_2 in five out of seven cities in period II were higher by 1–26% than period I. This recognizable increase in period II is likely to reflect the effect of increasing consumption rates in primary energy (e.g., petroleum and LNG). An examination of the seasonal trend of NO_2 consistently indicates the highest concentrations occurred during winter because of the combined effects of the anthropogenic emission and meteorological conditions. A health risk assessment of our data indicated that the NO_2 exposure (to adults, children, and infants) increased from period I to period II. Also, the long-term trends of NO_2 were analyzed based on the seasonal Mann–Kendall test and the Sen's slopes. It revealed that NO_2 levels of most cities had the linearly increasing trends during period I. However, decreasing trends appeared during period II to reflect the direct effect of implementation of administrative efforts including the fuel switching control policy.

1. Introduction

Nitrogen dioxide (NO_2) is formed from ground-level emissions related to the burning of fossil fuels in vehicles, power plants, industrial sources, and off-road equipment (EPA, 2011; Felix and Elliott, 2014). The NO_2 sources can be both primary (e.g., direct emissions from vehicle exhaust) and secondary (transformation via photochemical processes). Numerous publications have addressed the importance of primary NO_2 emissions from vehicles in urban areas (e.g., Anttila et al., 2011).

In the literature, increases in the NO_2/NO_x ratio from road traffic emissions are generally explained by the effects of (i) the increased use of diesel particulate filters (and oxidation catalysts) and (ii) the increased proportion of diesel cars in the automobile fleet (Mavroidis and Chaloulakou, 2011). The primary NO_2 emissions are considerably more important from diesel vehicles than from gasoline vehicles (Lenner et al., 1967; Carslaw and Beevers, 2005). The NO_2 exhausted from an idling engine is about four times higher than that from a car driven at a steady 40 km h^{-1} (Lenner et al., 1967). On major roads, the higher the vehicle speed, the lower the fraction of primary NO_2 emissions (by volume) (Carslaw and Beevers, 2005). They also recognized that a low vehicle speeds tend to result in a higher NO_2/NO_x emission ratio.

Ambient NO_2 concentration is mainly attributable to secondary processes through photochemical reactions in the atmosphere (Mavroidis and Chaloulakou, 2011). It is also known that nitrogen

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oxides (NO_x) consist of mixture of nitric oxide (NO) and NO_2, playing a major role in the formation of tropospheric ozone through a sequence of atmospheric chemical reactions (Altshuller, 1986; Finlayson-Pitts and Pitts, 1997). NO_2 is a strong oxidizing agent that reacts with OH to form corrosive nitric acid (HNO_3) which can directly contribute to the acidity of rainwater in the region (Waller et al., 2012). NO_2 air pollution affects human health by damaging the lungs and respiratory system (Pandey et al., 2005). Increased cardiopulmonary mortality was observed in people living near major roads in the Netherlands (Hoek et al., 2002). The relationship between human health and traffic-related NO_2, therefore, has been evaluated in many recent studies (Katsoulis et al., 2014).

The environmental significance of NO_2 in urban areas in Korea has been investigated in a number of our previous studies (Nguyen and Kim, 2006; Shon and Kim, 2011). In this paper, as an extension of our efforts to investigate major pollutants in the urban air, we analyzed the long-term trends of NO_2 using the datasets measured from seven major cities in S. Korea, Seoul (SL), Busan (BS), Daegu (DG), Incheon (IC), Gwangju (GJ), Daejeon (DJ), and Ulsan (UL) during the 1989–2010 period. For a simple comparison of NO_2 levels across two decadal periods, our data were divided arbitrarily into two periods of I (1989–1999) and II (2000–2010) to assess the difference in their temporal distribution patterns. Its environmental behavior is thus discussed in many diverse aspects based on both spatial and temporal (i.e., monthly, seasonal, and annual) analyses along with correlation analysis with relevant parameters such as criteria pollutants (SO_2, CO, O_3, TSP, and PM_{10}) and heavy metals (Pb, Cd, Cr, Cu, Mn, and Fe). In addition, a NO_2 health risk assessment was also carried out for adults, children, and infants for two decadal periods for each city.

2. Materials and methods

2.1. Site characteristics

In this study, we monitored the concentrations of NO_2 routinely from air quality monitoring (AQM) stations dispersed in seven major metropolitan areas in Korea (1989–2010). The number of individual urban background AQM stations operated in the seven cities increased gradually each year throughout the entire study period (i.e., from 21 stations (1989) to 89 stations (2010): Table 1S). The general geographical information and climate of the seven metropolitan cities (Fig. 1S) were described elsewhere (Ray and Kim, 2014). Most metropolitan cities in Korea have a high population density. Rural people seeking better employment opportunities in large industrial facilities near urban areas has accelerated the internal migration from rural to urban areas in S. Korea.

As S Korea experienced rapid economic growth, such effects are reflected in the statistical data. The population of the seven major cities represents about half of the national population (48.6 million) with the capital, Seoul of 9.79 million; these 7 large cities generated 44.1% of 2010 national GDP (Korean environmental statistics Environment Statistics Yearbook, 2012). The rapid growth in GDP and urban population possibly caused a large increase in the automobile fleet size together with a rapid increase in energy consumption. The number of automobiles in Seoul, representing more than half of the nationwide automobile fleet, increased from one million (in 1990) to more than two million (in 1995). By 2009, that figure dramatically grew to 10 million (Government of Seoul, http://www.wpro.who.int/environmental_health/documents/docs/SeoulReportonESHUT.pdf.). In particular, the population commuting daily in Seoul and nearby satellite cities (within a radius of 70 km from Central Seoul) exceeded 25 million. The daily traffic flow within the city was about 8 million vehicles/day (Kim et al., 2009).

Besides, many industrial complexes were constructed in vicinity of Seoul such as the Ansan industrial complex. Incheon has many different local emission sources, including seven industrial complexes, two seaports with ten wharfs, and one international airport (Geng et al., 2011). Another major industrial area established in Ulsan, the Hyundai industrial complex is the world's largest auto production complex and shipyard. The city has the second biggest petrochemicals complex in Korea (Jacobs, 2011). Daegu specializes in textile and electronics with simple industrial structures. Busan has the second largest population in Korea with 3.415 million (in 2010) and is Korea's gateway to the world's economy as the main port of trading various goods with other countries.

2.2. Data processing for temporal trend analysis

The original NO_2 data sets measured at the AQM stations are preprocessed and stored in the data management system operated by the Korean Ministry of the Environment (KMOE). All the initial data sets for criteria pollutants (e.g., SO_2, O_3, CO, TSP, and PM_{10}) were collected hourly from the monitoring stations (1998–2010), and then the data sets have been converted into the monthly data. In the same study period, heavy metals (e.g., Pb, Cd, Cr, Mn, and Fe) were also monitored monthly on a routine basis. To assess the factors affecting the distribution patterns of NO_2, all data in the whole period (1989–2010) were grouped into two data sets in period I (1989–1999) and period II (2000–2010) for further analysis. We analyzed the data for each city on several temporal scales (monthly, seasonal, and inter-annual intervals).

One of the most important objectives of air quality monitoring is to project long-term trends of the target pollutants. Because NO_2 concentrations were measured continuously at seven cities, we hypothesized that the temporal trends varied in a consistent manner across all study locations (i.e., cities) and across all seasons (i.e., months). To this end, we employed two Chi-square statistics tests (van Belle and Hughes, 1984). The homogeneity in a temporal trend is checked by the regional data (e.g., $n = 22 * 12$ for all SL data between 1989 and 2010) across all cities or by comparing the seasonal data (e.g., $n = 22 * 7$ for all January data between 1989 and 2010) across all months. If there was no commonality in the temporal trend across all cities, then individual trend tests of data for each seven cities are investigated. When there is no communality in the temporal trend both across all cities and across all months, then we need further analysis of data at each individual city over each month.

A non-parametric statistical test, namely Mann–Kendall (MK) statistics (Gilbert, 1987) is particularly useful to evaluate linear trends (either upward or downward) when data are positively correlated in time and space. Note that the null hypothesis of the MK test is set to exclude the existence of any (increasing or decreasing) long-term trend. Consequently, if the null hypothesis is rejected (at a specified significance level), then the linear slope calculated by the Sen's method (Gilbert, 1987) becomes significant. In this study, we arbitrarily set a significance level of 5%.

2.3. Health risk assessment

The average dose rates and health risk of adults, children (8–10 year olds), and infants were calculated using the concentration of NO_2 for all study period (1989–2010), period I (1989–1999), and period II (2000–2010) of seven cities. The daily dose rate of NO_2 was evaluated by following the procedures of Pandey et al. (2005),