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# Long-term trend of airborne particulate matter in Seoul, Korea from 2004 to 2013



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#### HIGHLIGHTS

- PM concentrations are monitored from the most urbanized location of Seoul Metropolitan area.
- Major factors regulating the PM levels over a decadal period are explored in a number of respects.
- The overall trend of PM confirms the potent role of legislation efforts to improve the air quality.

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#### ABSTRACT

In this study, the pollution status of particulate matter (PM) in ambient air was investigated based on the concentration data of three important PM fractions (PM<sub>2.5</sub>, PM<sub>10</sub>, and TSP) measured from a central area in Seoul, Korea during the period from 2004 to 2013. The mean concentrations of each fraction measured for the entire study period were found to be 26.6  $\pm$  2.59, 54.0  $\pm$  15.0, and 75.3  $\pm$  16.6  $\mu g$  m $^{-3}$ , respectively. The seasonal mean of PM<sub>2.5</sub> varied in the range of 22.9  $\pm$  7.10 (fall) to 30.2  $\pm$  7.58  $\mu g$  m $^{-3}$  (winter). In contrast, PM<sub>10</sub> and TSP showed a summer minimum (40.1  $\pm$  12.6 and 55.6  $\pm$  17.8  $\mu g$  m $^{-3}$ , respectively) and a spring maximum (67.1  $\pm$  16.7 and 93.7  $\pm$  21.1  $\mu g$  m $^{-3}$ , respectively). The contribution of regional or long-range transport to the observed PM levels in the study area, if estimated by comparison to the data of the regional background area, was found to explain up to 72% of its input. The long-term trend of PM indicated a gradual decreasing pattern over a 10 year period, although that of PM<sub>2.5</sub> was rather complicated to interpret in the recent years. The overall results of our study nonetheless confirm the potent role of legislation efforts put consistently to improve the air quality through the years.

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#### 1. Introduction

Particulate matter (PM) is a pollutant of great environmental concern due to its association with increased mortality (from non-accidental and cause-specific diseases) (Samoli et al., 2005; Eftim et al., 2008; Brook et al., 2010). There have been numerous efforts to demonstrate a strong relationship between mortality (and various morbidity outcomes like hospital admission and pulmonary function) and PM (Zanobetti and Schwartz, 2005; Dominici et al., 2007; Betha et al., 2013). PM is also known to exert significant effects on the alteration of the earth's radiative balance (Forster et al., 2007;

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Yin and Harrison, 2008). However, the mechanism and the extent of such an association still remain unknown for further investigation (Dockery and Stone, 2007).

Based on the aerodynamic diameter of particles, PMs are classified into  $PM_{2.5}$  (PM with an aerodynamic diameter  $< 2.5 \mu m$ ),  $PM_{10}$  (PM with an aerodynamic diameter  $< 10 \mu m$ ), and TSP (total suspended particles) (Kleeman et al., 2000; Kim et al., 2010). PM exists as a complex mixture of extremely small particles and liquid droplets including acids (such as nitrates and sulfates), metals, organic chemicals, etc. The extent of its pollution in the atmosphere can thus be affected by a number of variables including the number, size, shape, surface area, chemical composition, solubility, and origin (Pope and Dockery, 2006; Saliba et al., 2010).

Because PM is emitted into the atmosphere by the combined effects of anthropogenic and natural sources, the physical and chemical patterns of its emissions may vary considerably by the factors controlling their source characteristics (Kim et al., 2003; Alastuey et al., 2006). A large-scale dust storm frequently occurring in association with strong winds and exposed soils (namely Asian Dust (AD)) is a common phenomenon in China and surrounding countries from winter to spring (Natsagdorj et al., 2003; Lee et al., 2006; Hao et al., 2007). As such, AD events have the great potential to alter the PM concentrations and the air quality (Liu et al., 2006; Chun and Lim, 2004). Because air pollution has become a very common phenomenon, the study of PM pollution and its associated impacts is now treated as a very important component of air quality management from many countries in the world (Gong et al., 2006).

Korea has experienced rapid and advanced industrialization (Oh and Lee, 2013). Such advancement has contributed greatly to the deterioration of its environmental conditions including ambient air quality (Lim et al., 2010; Gummeneni et al., 2011). To improve air quality and public health, legislation has been strengthened against some specific pollutants (including PM) (Neuberger et al., 2004; Le Tertre et al., 2005).

In this study, the analysis of PM<sub>2.5</sub>, PM<sub>10</sub>, and TSP was performed using data sets collected concurrently from Yongsan, which represents the urban center of the capital city, Seoul, Korea. Consequently, our study aims to provide the general picture of PM pollution in the most urbanized location of Korea over the most recent decadal period. The analysis of PM pollution in the study area was investigated by focusing on the air mass transfer pattern for an annual period of 2013 (Vellingiri et al., 2015). Here the patterns of the PM pollution were explored for an extended period from 2004 to 2013 by investigating the behavior of three different PM fractions after grouping the data into diverse temporal (daily, monthly, seasonal, and annual) scales. In addition, we also examined the factors affecting the behavior of airborne PM in relation to relevant environmental parameters.

#### 2. Materials and methods

#### 2.1. Site characteristics of the study area

In this study, to investigate the temporal trends of airborne PM at various intervals, we analyzed the concentrations of PM<sub>2.5</sub>, PM<sub>10</sub>, and TSP data measured from Yongsan (YS), Seoul, Korea (37°32′18″N and 126°57′56″E) between 2004 and 2013. The study site, YS is located on the north of the Han River under the shadow of Seoul Tower and belongs to one of the most densely populated districts of Seoul with about 238,920 people as of May 2014 (http:// en.wikipedia.org/wiki/Yongsan\_District). Geographical location of the study site is presented in Fig. S1. The site is well known for some landmark buildings like Yongsan Station, the sprawling Yongsan Electronics Market, Haebangchon, the Itaewon commercial district and Yongsan Garrison (a large United States military base in the heart of Seoul). The headquarters of some well known KOSPI 200 companies including Amore Pacific, Orion Confectionery, Cheil Worldwide, and Hyundai Development Company are also based in Yongsan due to its prominent industrial importance. Further, the Ministry of National Defense in this district also adds to its administrative significance.

#### 2.2. Data processing

The concentration data of different PM fractions have been collected routinely along with other criteria pollutants (including CO, NO<sub>2</sub>, SO<sub>2</sub>, and O<sub>3</sub>) from both regional (urban) background and roadside stations dispersed all across seven major metropolitan cities and a number of small scale cities in Korea (Kim et al., 2010; Kim and Shon, 2011). As our study site, YS belongs to one of these

network, all of these monitoring tasks have been conducted by following the routine protocol for the AIR QUALITY monitoring operation guide and regulations managed by the Korean Ministry of Environment (KMOE).

The hourly data of three PM fractions at the study site were measured continuously using ambient dust monitor (FH 62 C14, Thermo Electron Corporation, USA). This monitoring system is built to operate by the principle of beta attenuation (using a  $C_{14}$  source) through particles collected on a movable filter tape. The measuring position is continuously fed with a new section of the filter tape. Then, ambient air is pulled through the inlet and the sample tube to induce the deposition of dust on the filter through which the beta beam passes. The intensity of the beta beam expressed as count rate from the detector is inversely proportional to the dust mass loaded on the filter. The minimum detection limit for PM<sub>2.5</sub>, PM<sub>10</sub>, and TSP are  $<1 \mu g m^{-3}$  with the precision of  $\pm 2 \mu g m^{-3}$ . The output data were derived from the serial interface RS 232 using 4-20 mA or 0-10 V analog output of concentration (µg m<sup>-3</sup>). The full scale management of the QA procedure is conducted routinely at Quarterly intervals unless there are special needs for the management. The detailed instrumental set up is described in Table S1. For the sake of simplicity, the raw hourly concentrations of each PM fraction were converted into the daily data sets. Hence, all of our analyses were made using these daily data either as they are or after being sorted into month or seasonal groups.

The concentrations of other selected pollutants (e.g., CH<sub>4</sub>, SO<sub>2</sub>, O<sub>3</sub>, and CO) were also measured using HA-675, Hydrocarbon Analyzer; SA-633, Sulfur Dioxides Analyzer; OA-683 Ozone Analyzer (all three by KIMOTO, Japan); and ZRF, Gas Analyzer (FUJI, Japan) instruments, respectively. The wind speed data were measured using the high performance wind sensor "Wind Monitor" (Model WM0513, R.M. Yong Company, USA). The temperature and humidity were measured using high-altitude meteorological mast (MHP45A, VAISALA Co., Finland). Besides, the ultraviolet (UV) and solar radiation (SR) data were measured using pyranometer (LI200SZ, LI-COR Biosciences, USA).

Theoretically, the total count for the daily data of PM and relevant parameters should reach 3653 for the 10 year study period. However, in practice some data were missing, i.e., acquisition of no data due to the malfunctioning of the measurement system or the lack of representativeness; we omitted the daily data, if less than 2/3 (n<16) of hourly data were collected for a given day. The maximum number of total daily data was thus counted as 3544 in this study.

To examine the factors affecting the behavior of airborne PM, the effect of regional or long-range transport (LRT) on observed PM levels was evaluated indirectly at the target site (YS, the urban site with low elevation) by adopting a simple regression approach that has been previously employed (Karppinen et al., 2004; Timonen et al., 2013). In general, increases in PM concentrations due to such transport can frequently occur in a free troposphere (Timonen et al., 2013). It can thus be assumed that PM observed at background sites with relatively high elevation should be dominated by regional or LRT sources (e.g., from China), because the local sources are placed at a much lower elevation (Jaffe et al., 2003). It is assumed further that local PM represents a fraction that does not correlate with the regional or LRT PM. To this end, a background monitoring site located near the boundary layer (i.e., Mt. Bukhan site located 13.52 km north from Yongsan) was selected. Then, a regression analysis was carried out using PM<sub>2.5</sub> data from the two sites (YS vs Mt. Bukhan). The regression analysis was also made using the data for 7 years (2006 and 2008–2013) due to the limited availability of data at the background site. Note that PM<sub>2.5</sub> data at Mt. Bukhan site was obtained from the official website of the Seoul Metropolitan Government (http://env.seoul.go.kr/statistics\_air).

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