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Trend of Air Quality in Seoul: Policy and Science

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ABSTRACT

The trend of air pollutant concentrations in the Seoul Metropolitan Area (SMA)—particularly the city of Seoul—in the Republic of Korea, is shown and analyzed along with applied policy; furthermore, the remaining challenges are identified, and the direction of future research is discussed. The policies adopted from developed countries, notably, direct emission control measures, such as limiting the sulfur content in fuel and tightening emission standards, have been successful in reducing primary air pollutants, e.g., carbon monoxide, sulfur dioxide, and lead; however, these policies have not been effective in controlling the increased number of emission sources and secondary air pollutants, such as particulate matter with an aerodynamic diameter less than or equal to a nominal 2.5 μm ($PM_{2.5}$), and ozone. To develop effective control policies on air pollution, the following actions are recommended: (1) creating a reliable emission inventory; (2) reducing uncertainties about the regional contribution to the air quality in Seoul; and (3) understanding the major chemical pathways of ozone and secondary aerosols. Suggestions for accomplishing these goals in future research are also provided.

Keywords: Seoul; Air quality management policy; Trend of air pollutants; Secondary air pollutants; International cooperation.

INTRODUCTION

Generally, in an urban area, due to high population density along with concentrated economical activities, a huge amount of energy is consumed and, thus, a huge amount of environmental burden including air pollutants is emitted. Thus, in urban areas, there exist several environmental problems including air quality problems. Urban air pollution is a major environmental problem in the world. Several international and country specific activities have been carried out to alleviate the problem. For example, the United Nations Environmental Programme (UNEP) and the World Health Organization (WHO) have started a global environment monitoring system, the urban air pollution monitoring network (GEMS/Air) in 1974 and it was reported that air pollution is widespread across the urban areas in the world, especially in developing countries (Mage *et al.*, 1996).

To improve air quality in urban areas, several measures have been developed and implemented. There are several common factors that have caused air pollution problems in one urban area. It is widely thought that the trends in air quality in different cities show similar pattern. There are also, however, some factors that are specific to each urban area. When these factors are all understood, appropriate control strategies can be developed and implemented.

Seoul and the Seoul Metropolitan Area (SMA) take up only 0.6% and 11.8% of the total national land area of the Republic of Korea (Korea) as shown in Fig. 1, respectively (City of Seoul, 2018; KOSTAT, 2018). However, they account for 19.7% and 49.5% of the total population in 2016, and 14% and 44% of national total registered vehicles in 2017, respectively (City of Seoul, 2018; KOSTAT, 2018). Such densely populated situation makes the management of urban air quality very difficult. For example, the concentration of $PM_{2.5}$ in Seoul marked 1.7–2.7 times higher than those in other major cities, such as Tokyo, Paris, and New York in 2015 (City of Seoul, 2018). Against these serious challenges, various measures have been implemented such as the Special Measures for Metropolitan Air Quality Improvement with varying degree of success.

In this work, (1) the temporal trend of air quality, especially, that of ambient particles in Seoul is presented along with the policy measures and (2) remaining challenges and further research directions to efficiently reduce air pollution level in Seoul are discussed.

AIR QUALITY MANAGEMENT FOR SEOUL AND THE TREND OF THE AIR POLLUTANTS' LEVEL

Policy

In Seoul, the capital of Korea, the concentration of

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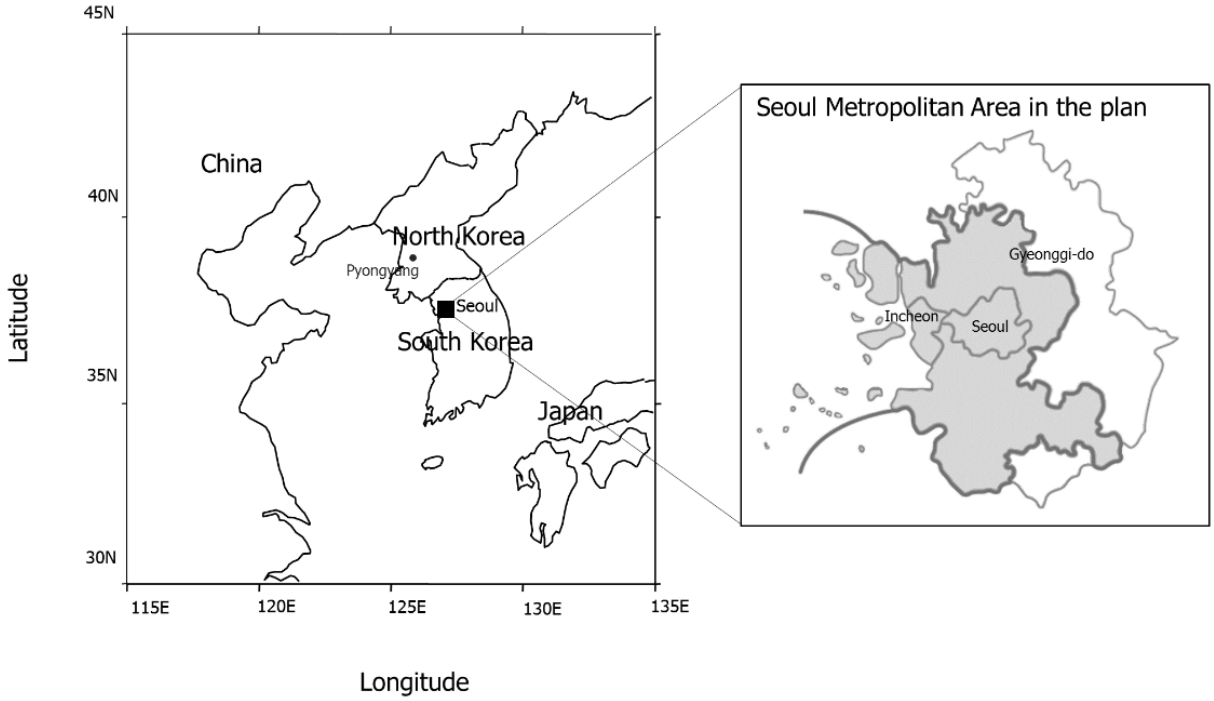

Fig. 1. Map of the Republic of Korea and the Seoul Metropolitan Area (SMA) (MOE, 2007).

sulfur dioxide (SO_2) was among the highest in the world until the 1980s (Mage *et al.*, 1996) and, thus, with high public discontent. In Korea, the first national air quality standard was established in 1978 for sulfur dioxide (SO_2) and then it has been expanded to include 8 air pollutants as in 2017 as shown in Table 1.

Since high SO_2 level in an urban area is mainly due to the use of high sulfur content fuels such as coal and heavy oils, the major air quality management strategies in the 1980s and 1990s for Seoul were regulations on the sulfur content in petroleum and on the use of solid fuels as shown in Table 2. The decreasing trend of SO_2 is shown in Fig. 2 and that was closely related with the change of fuel types as shown in Table 2. The Ministry of Environment (MOE) in Korea has limited the use of solid fuel and heavy oils for heating and cooking in the SMA since 1985 and strongly enforced the rule since 1995 (MOE, 2005). Thus, the amounts of coal consumption for residential, industrial, and commercial sectors at Seoul have been rapidly decreased since 1990 and the usage of coal has been negligible since 1995. The amount of natural gas consumption at Seoul has been rapidly increased since 1990 (Kim, 2006).

Along with SO_2, the concentrations of other primary air pollutants such as carbon monoxide (CO), lead (Pb), and total suspended particles (TSP) have also decreased during the same period. As shown in Figs. 2 and 4, from 1999 to 2002 the concentrations of TSP and PM_{10} have increased while the levels of SO_2, CO, and Pb have decreased as shown in Fig. 2. In Fig. 3, the trends of emissions of TSP, PM_{10}, and CO are shown. Since the official emission inventory of Korea, the Clean Air Policy Support System (CAPSS) has been established from 1999, only the emission data from 1999 are comparable to each other. While the emissions of CO and SO_2 (shown in Fig. 5) decreased during the period, those of TSP and PM_{10} have increased, and, thus the ambient concentrations. Also note that the concentrations of these primary air pollutants have not decreased significantly from the early 2000s. This trend is thought to be caused by the increase of the emission amounts due to the increase of the number of emission sources such as vehicles and domestic boilers. For example, the number of vehicles in Seoul has increased from 0.2 million in 1980, 1.2 million in 1990, and to 2.4 million in 2000 (SI, 2003). Thus, the total amount of air pollutants' emission has increased even with the tighter emission standard for each source. This increasing emission trend is also shown for the oxides of nitrogen in Fig. 6 and VOCs in Fig. 10, respectively.

To counter-measure this trend, the Korean Government has enacted the "Special Act on the Improvement of Air Quality in Seoul Metropolitan Area" in 2003 (enforced in 2005) and based on the Act, the "1st Seoul Metropolitan Air Quality Control Master Plan" was executed between 2005 and 2014. The goals of the 1st Plan were to accomplish the annual average concentrations of PM_{10} and NO_2 target values of 40 μg m^{-3} and 22 ppb in Seoul, respectively. Major directions of the Special Act are shown in Table 2. Most of the budget (more than 90%) went to the reduction of the emissions from diesel vehicles.

During the 1st Plan period, total investment of ~3 trillion Won (~2.7 billion USD) was made. The 2nd Plan is in operation from 2015 to 2024. In addition to PM_{10} and NO_2, $PM_{2.5}$ and ozone (O_3) are included as target species. The target values are 30 μg m^{-3} for PM_{10}, 20 μg m^{-3} for $PM_{2.5}$, 21 ppb for NO_2, and 60 ppb for ozone (8 hrs) in Seoul by 2024, respectively (MOE, 2016).

Table 1. Evolution of the ambient air quality standard of the criteria air pollutants in Korea.

Year		1983	1991	1993	2001	2007	2010
Sulfur Dioxide (SO_2, ppm)		0.05/year* 0.15/day*	S	0.03/year** 0.14/day** 0.25/hour**	0.02/year 0.05/day 0.15/hour	S	S
Carbon Monoxide (CO, ppm)		8/month 20/8hour	S	9/8hours 25/hour	S	S	S
Nitrogen Dioxide (NO_2, ppm)		0.05/year 0.15/hour	S	0.05/year 0.08/day 0.15/hour	S	0.03/year 0.06/day 0.1/hour	S
Particulate Matters ($\mu g\ m^{-3}$)	TSP	150/year 300/day	S	S	-	-	-
	PM_{10}	-	-	80/year** 150/day**	70/year 150/day	50/year 100/day	S
	$PM_{2.5}$	-	-	-	-	-	25/year*** 50/day***
Ozone (O_3, ppm)		0.02/year 0.1/hour	S	0.06/8 hours 0.1/hour	S	S	S
Lead (Pb, $\mu g\ m^{-3}$)		-	1.5/3 months	S	0.5/year	S	S
Benzene ($\mu g\ m^{-3}$)		-	-	-	-	-	5/year
Total hydrocarbon (ppm)		3/year 10/hour	S	-	-	-	-

* From 1978; ** From 1995; *** From 2015. 'S' means the same as the existing value and '-' means no standard.
Standards for hour base values: 99.9 percentile value of one-hour average value should not exceed the standard value and 99 percentile value of eight- or 24-hour value would not exceed the standard value.
Data are from the National Institute of Environmental Research (NIER).

Table 2. Major policy against air pollution implemented in Seoul and Korea.

Policy	Year	Item	Target area
Regulation on the sulfur content in petroleum	(1) 1981 (2) 1993 (3) 1996 (4) 2001 (5) 2012	(1) 1.6% for heavy oil, 0.4% for diesel (2) 1.0% for heavy oil, 0.2% for diesel (3) Same as (2) (4) 0.3% for heavy oil, 0.1% for diesel	(1), (2), (4) Seoul and major cities (3) Small cities (5) Small cities for heavy oil and nationally for diesel
Regulation on solid fuels	(1) 1985 (2) 2003	(1) Restriction of the use of solid fuels such as coal and wood (2) Ban the use of solid fuels	(1) SMA and large cities (2) Seoul
Use of clean fuels (LNG and LPG)	(1)1988 (2) 1991	(1), (2) Mandatory use of clean fuels	(1) Seoul (2) SMA
Regulation of leaded gasoline	1993	No sales of leaded gasoline	Korea
Designation of the air quality control area	1997	(1) Compulsory installation of VOC inhibition and prevention facilities (2) Strict inspection of exhaust gases from vehicles in operation, and introduction of low-pollution vehicles	Seoul
Special act on the Improvement of Air Quality in Seoul Metropolitan Area	2003	(1) To reform atmospheric administration system (2) To introduce emission cap (cap and trade) system (3) To take measures to reduce atmospheric pollution from vehicles (4) To strengthen volatile organic compounds management (5) To establish eco-friendly management of city and energy	SMA

Data are from Ministry of Environment (MOE), Korea.

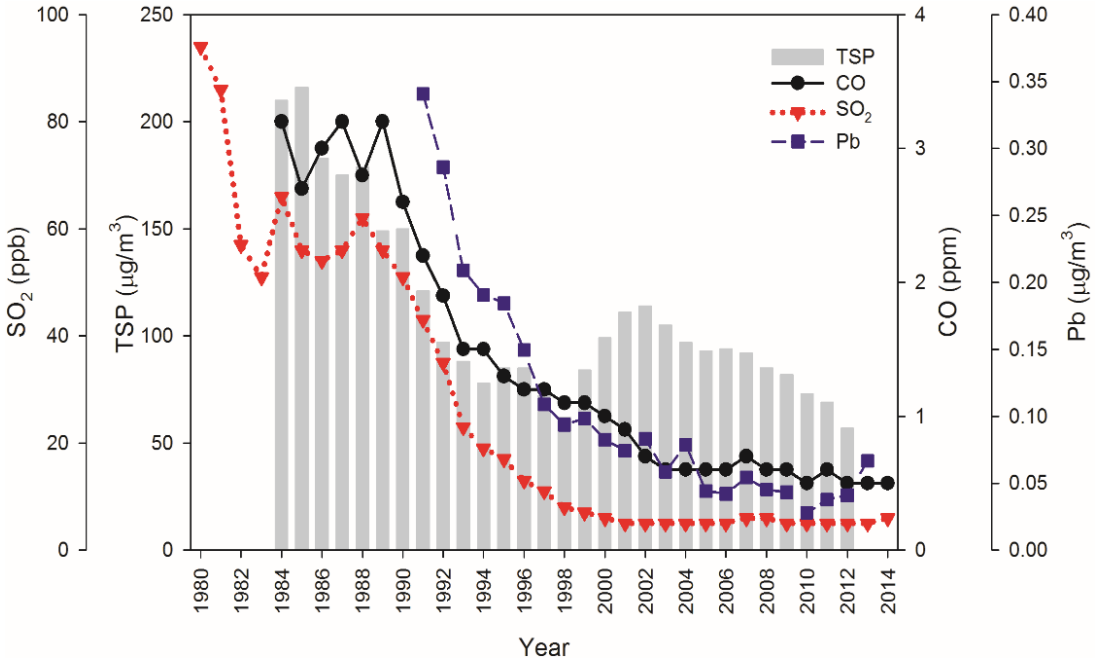

Fig. 2. Trend of the concentration of primary air pollutants in Seoul between 1980 and 2014. Data are from NIER and City of Seoul.

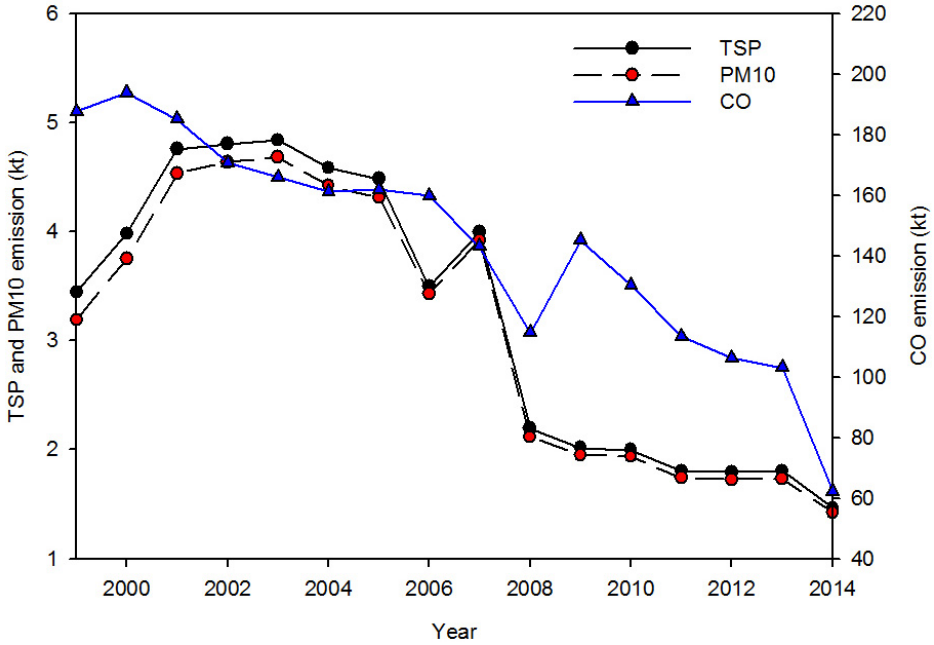

Fig. 3. Trends of the emission amounts of TSP, PM_{10}, and CO in Seoul between 1999 and 2014. Data are from NIER.

Trend of the Concentration of Air Pollutants

Major chemical species in $PM_{2.5}$ observed in Seoul have been carbonaceous species and inorganic ions. In this section, trend of the levels of inorganic and carbonaceous species and volatile organic compounds, ozone, and visibility is discussed.

Mass Concentrations of $PM_{2.5}$ and PM_{10}

The concentrations of $PM_{2.5}$ and PM_{10} in Seoul from 2003 are generally decreasing as shown in Fig. 4. The main reason for this decreasing trend might be the reduction of the emissions of air pollutants in Seoul and the SMA, though a study suggested that meteorological conditions have also contributed to the decreasing trend (Lim *et al.*, 2012). According to the MOE, emissions of PM_{10}, NO_x, SO_x, and Volatile Organi Compounds (VOCs) in Seoul and the SMA decreased 64%, 45%, 55%, and 28%, and 33%, 30%, 40%, and 9% in 2014 comparing to those in 2007, respectively (MOE, 2018).

In 2013, the concentrations of both PMs had increased

and it was suggested that this increase was mainly due to the change of the climate conditions, not the increase of the emissions of air pollutants (Kim *et al.*, 2017).

Sulfate

The concentration of sulfate in Seoul has decreased from the 1980s (more than 15 μg m^{-3}) and 1990s (around 10 μg m^{-3}) and stabilized from the 2000s (around 6–7 μg m^{-3}) as shown in Fig. 5. In Fig. 5, the variations of the emission amount of sulfur oxides (SO_x) and the ambient concentration of SO_2 are also shown. It is evident that the emission amounts in the early 2010s have decreased

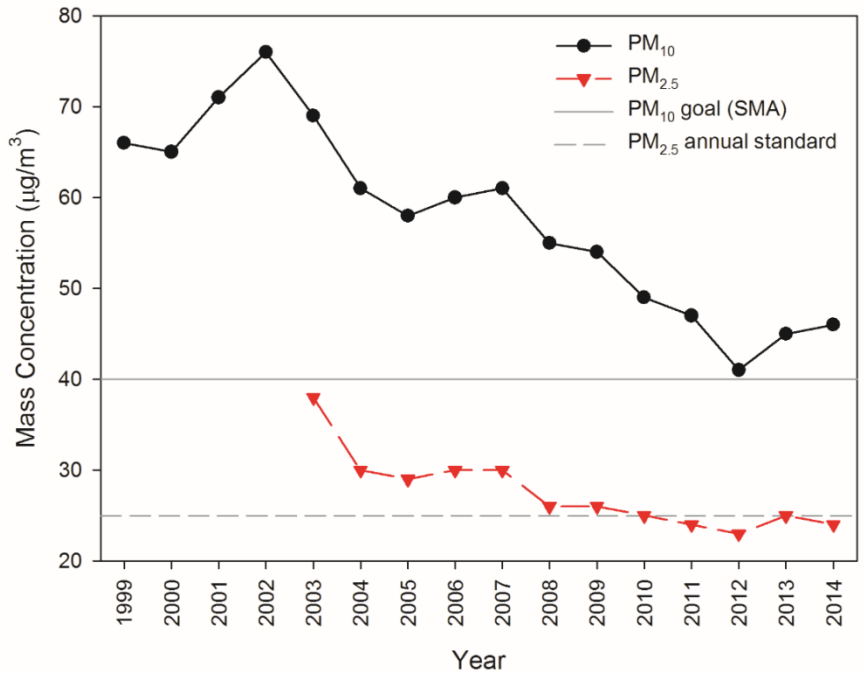

Fig. 4. Trend of the annual average concentrations of $PM_{2.5}$ and PM_{10} in Seoul. Data are from NIER and City of Seoul.

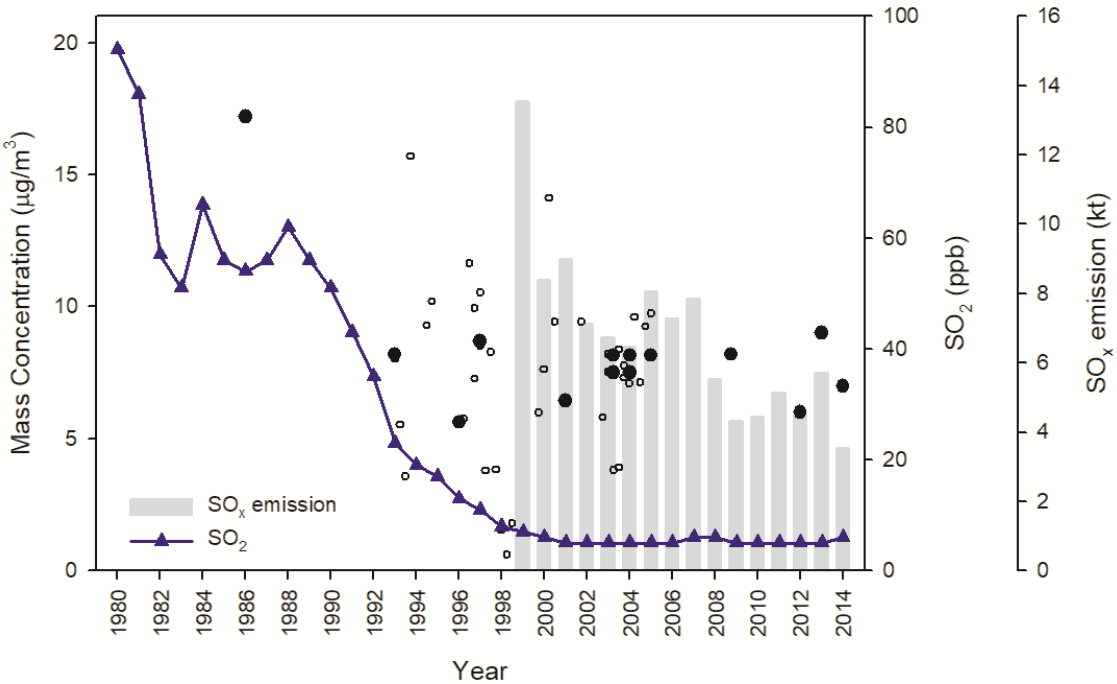

Fig. 5. Trend of the concentration of sulfur dioxide, sulfate in $PM_{2.5}$, and emission amount of sulfur oxides in Seoul between 1980 and 2014. Filled symbol is the yearly average concentration and open symbol is shorter period concentration data. Sulfate measurement data before 2012 are from Han and Kim (2015) and data from 2013 are from NIER. Sulfur dioxide and emission data are from NIER and City of Seoul.

noticeably compared to those in the 2000s. This trend is in contrast with the observed sulfate and SO_2 concentrations. Thus, it is suspected that the sulfate concentration in Seoul is influenced both by transport and generation over Seoul mostly from SO_2 emitted from outside of the SMA.

Modeling results by using the CMAQ have generally underestimated the concentration of sulfate in Seoul. Since SO_x emission estimated for both China and Korea are rather reasonable, it is suspected that the conversion process from sulfur dioxide to sulfate be underestimated. For example, He *et al.* (2014) reported that with mineral dust and NO_x, the conversion rate of SO_2 to sulfate increased in Beijing.

Nitrate and Ammonium

As shown in Fig. 6, the emission amount of the oxides of nitrogen (NO_x = NO + NO_2) has decreased about 40% from 1999 and 2014. Also, the concentration of NO_x has decreased significantly, about 30% during the same period. However, the concentration of NO_2, is almost the same. There are speculations on the cause(s) on this phenomenon.

One possibility is that the emission inventory for NO_x be underestimated. It is certain that the NO_x emission inventory is underestimated due to several reasons, one of them is the underestimation of the NO_x emissions from diesel vehicles. However, this hypothesis could not explain the decreasing trend of the ambient NO_x concentration. Furthermore, based on the inverse modeling result, it seemed that the emission inventory in Korea up to the middle of 2000s was reasonably good (Kim *et al.*, 2013b).

Another possibility is that the installation of the diesel vehicle emission control equipment such as diesel particulate filter (DPF) and diesel oxidation filter (DOC) might increase the oxidation state of the emitted NO_x from a diesel engine and, thus, increase the fraction of NO_2. The same phenomenon has been observed in European countries where the fraction of diesel vehicles has been high. However, it was found out that the difference of the NO_2/NO_x ratios at the roadside monitoring stations and at the urban monitoring stations in Seoul was small (SI, 2011). It is likely that the installation of the control equipment has contributed somewhat but not the major factor for the phenomenon.

Third hypothesis is that the ambient air in Seoul has become more oxidative and, thus, the conversion from NO to NO_2 has become faster. Similar speculation has been made based on the ozone concentration trend analysis which will be discussed later in this section.

The trend of the ammonium concentration is similar to that of nitrate, increasing trend as shown in Fig. 7. Ammonium is a basic ionic species neutralizing acidic ions such as sulfate and nitrate. Since sulfate concentration is not changing much, the ammonium concentration is critically related with nitrate concentration. Unfortunately, ammonia concentration data has not been routinely reported in Seoul.

Carbonaceous Species (OC and EC) and Volatile Organic Compounds (VOCs)

The concentrations of organic carbon (OC) and elemental carbon (EC) are shown in Figs. 8 and 9, respectively. OC is an important component in aerosol mass concentration and OC and EC affect several areas of atmospheric and

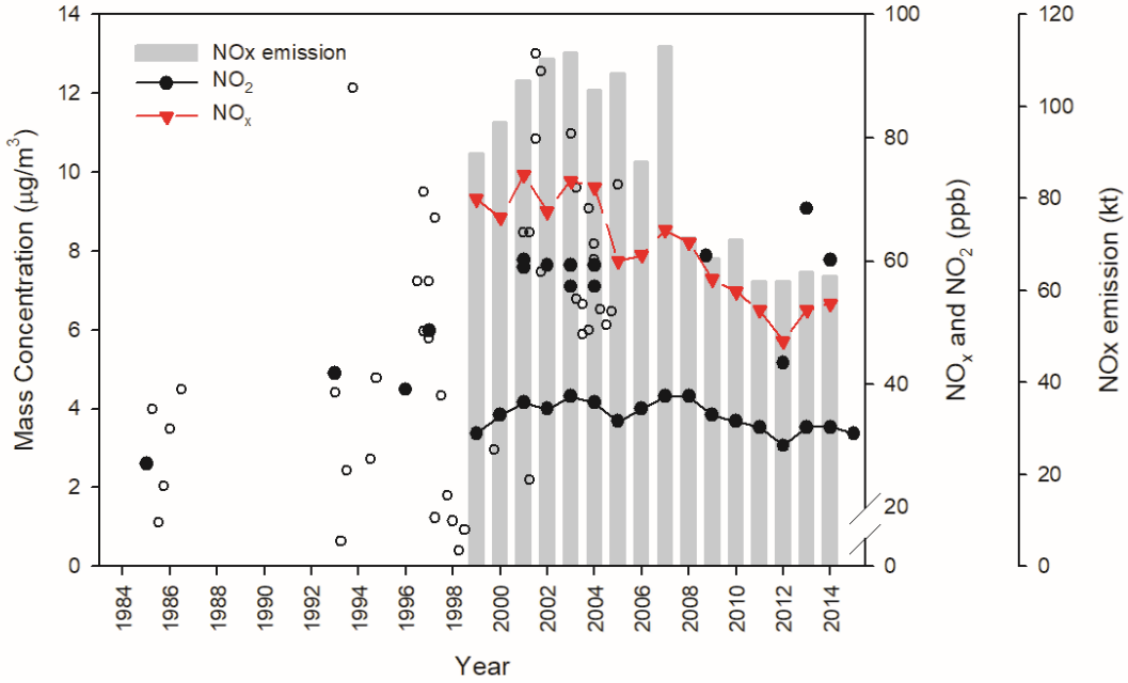

Fig. 6. Trend of the concentration of the oxides of nitrogen (NO_x = NO + NO_2), nitrogen dioxide (NO_2), nitrate in $PM_{2.5}$, and emission amount of the oxides of nitrogen in Seoul between 1985 and 2015. Filled symbol is the yearly average concentration and open symbol is shorter period concentration data. Nitrate measurement data before 2012 are from Han and Kim (2015) and data from 2013 are from NIER. Sulfur dioxide and emission data are from NIER and City of Seoul.

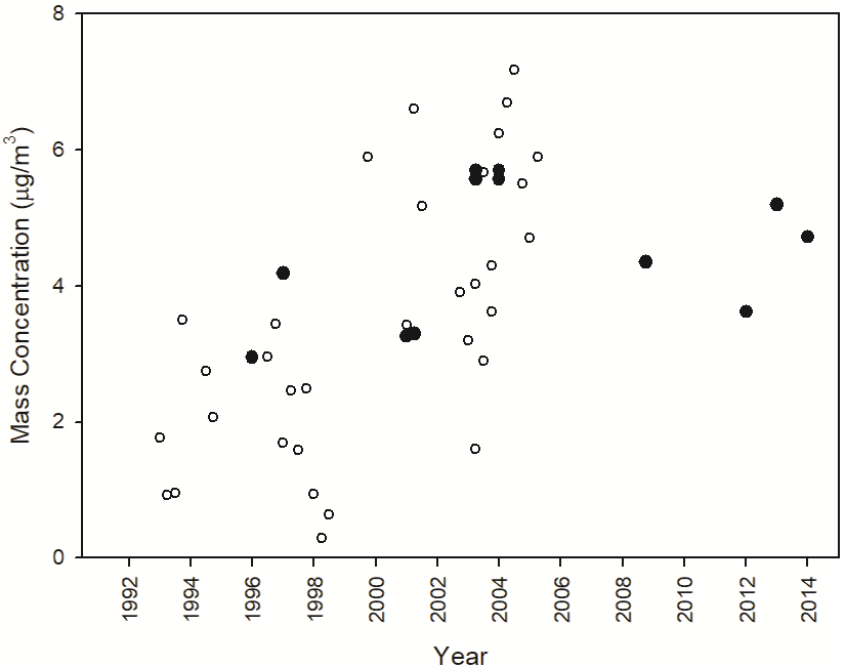

Fig. 7. Trend of the concentration of ammonium in $PM_{2.5}$ in Seoul between 1993 and 2014. Filled symbol is the yearly average concentration and open symbol is shorter period concentration data. Measurement data before 2012 are from Han and Kim (2015) and data from 2013 are from NIER.

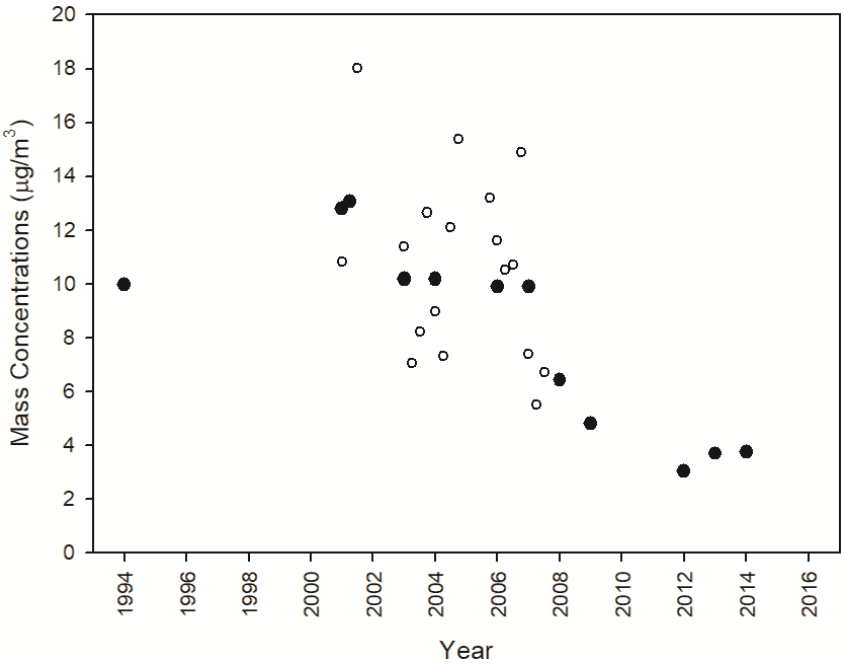

Fig. 8. Trend of the concentration of organic carbon (OC) in $PM_{2.5}$ in Seoul between 1994 and 2014. Filled symbol is the yearly average concentration and open symbol is shorter period concentration data. Measurement data before 2012 are from Han and Kim (2015) and data from 2013 are from NIER.

global environment (Kanakidou *et al.*, 2005). Both OC and EC concentrations decreased significantly during last 2 decades. The decreasing trend of EC during the 2000s was mainly due to the installation of DPFs and DOCs to diesel vehicles. The decreasing trend of OC was due to various government policy measures against primary and secondary organic carbon. Secondary OCs are generated from the reaction of VOCs.

VOCs have been analyzed using adsorption tubes for sampling and gas chromatography for analysis in Korea for aromatic compounds in the 1980s and early 1990s. The first comprehensive identification and quantification of the

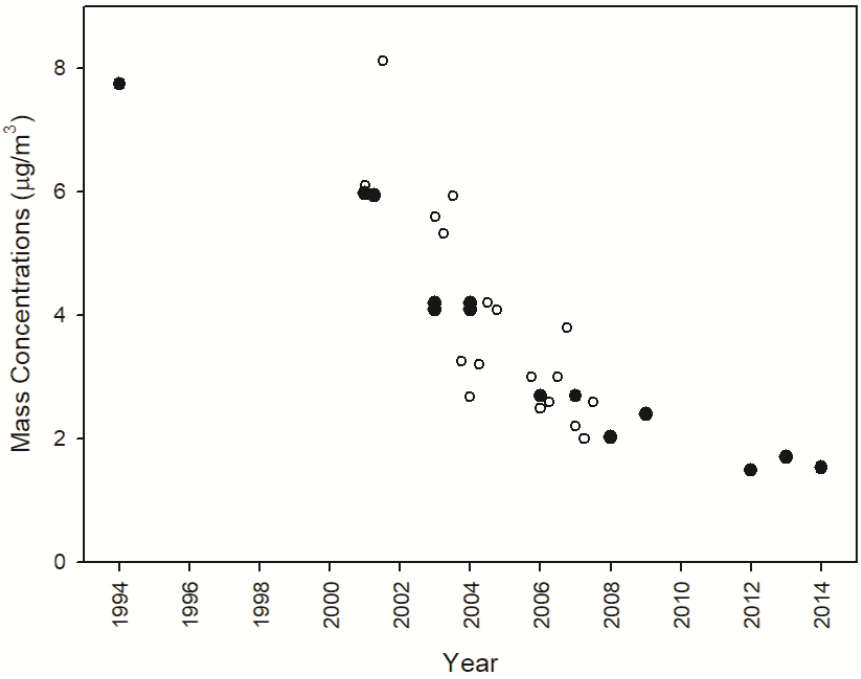

Fig. 9. Trend of the concentration of elemental carbon (EC) in $PM_{2.5}$ in Seoul between 1994 and 2014. Filled symbol is the yearly average concentration and open symbol is shorter period concentration data. Measurement data before 2012 are from Han and Kim (2015) and data from 2013 are from NIER.

VOCs in Seoul following the U.S. EPA method TO-14 (U.S. EPA, 1988) was carried out in the later 1990s. Na and Kim (2001) have analyzed 70 compounds from the SUMMA canister samples collected on every sixth day between August 1998 and July 1999. The mean concentration was 248.3 ppbC with the highest annual mean concentration of 7.8 ppb for propane, followed by 6.4 ppb of toluene, 5.9 ppb of ethylene, and 3.8 ppb of ethane. Alkanes provided the largest contribution to the total VOCs on ppbC base, varying from about 36–52%, followed by aromatics, accounting for 28–46% of the total VOCs. They suggested that in addition to vehicular exhaust and solvent usage, fugitive emissions from natural gas and Liquefied Petroleum Gas (LPG) might be important sources.

MOE has established the Photochemical Assessment Monitoring Station (PAMS) since 2002 to monitor the concentrations of the ozone precursors, VOCs and NO_x, responding to high ozone phenomenon (Shin *et al.*, 2013a). Bulgwang site is one of them and designated as the representative site for Seoul. Fig. 10 shows the annual VOCs emissions in kilo-tonne (kt) and the annual average concentration of total VOCs (55 compounds) in ppbC in Seoul. Between 2007 and 2014, trends of concentration and emissions of VOCs were similar. The total VOCs concentration decreased from 113.27 to 83.83 ppbC, a 26% decrease and the emissions of VOCs in Seoul and the SMA during the same period decreased 28% and 9% between 2007 and 2014, respectively (MOE, 2018).

Shin *et al.* (2013b) reported based on the Bulgwang data between 2004 and 2008 that the aromatics except benzene showed high concentrations in daytime since they are mainly emitted from solvent usage. On the other hand, alkanes, low carbon alkenes, and benzene showed high concentrations in morning and evening around commuting hours suggesting vehicular sources. The weekend effect was observed for these compounds only for Sundays.

There have been a few studies on the major emission sources of VOCs in Seoul. Na and Kim (2007) had applied the Chemical Mass Balance (CMB) Model including loss processes to the VOCs measurement data in September 1998 with the source profiles measured for Seoul (Na *et al.*, 2004). They found that (1) including loss processes would not change the relative importance of the major contributors though giving better modeling performance and (2) vehicular exhaust contributed about 52% followed by solvent usage (26%), gasoline evaporation (15%), LPG and natural gas (7%). Similar result was obtained from a Positive Matrix Factorization (PMF) modeling study (SI, 2012). They have applied the U.S. EPA PMF v3.0 model to the measurement data at Gui (residential area) and Guro (industrial area) between 2009 and 2010. These two sites are the intensive monitoring stations operated by city of Seoul and comparable to the Bulgwang site operated by MOE, analyzing 55 compounds. The modeling results for both sites were different due to their emission characteristics. Still, the contributions from motor vehicles were higher (44 and 48%) than solvent usage (37 and 31%) at Gui and Guro, respectively. As shown in Table 3, on the contrary, the official VOCs emission inventory of Seoul in 2009 showed higher contribution from solvent usage (69%) than fossil fuel combustion including mobile sources, energy transportation and storage (30%) (NIER, 2012).

Shin *et al.* (2013b) carried out a Principal Component Analysis (PCA) to identify the major sources of the VOCs

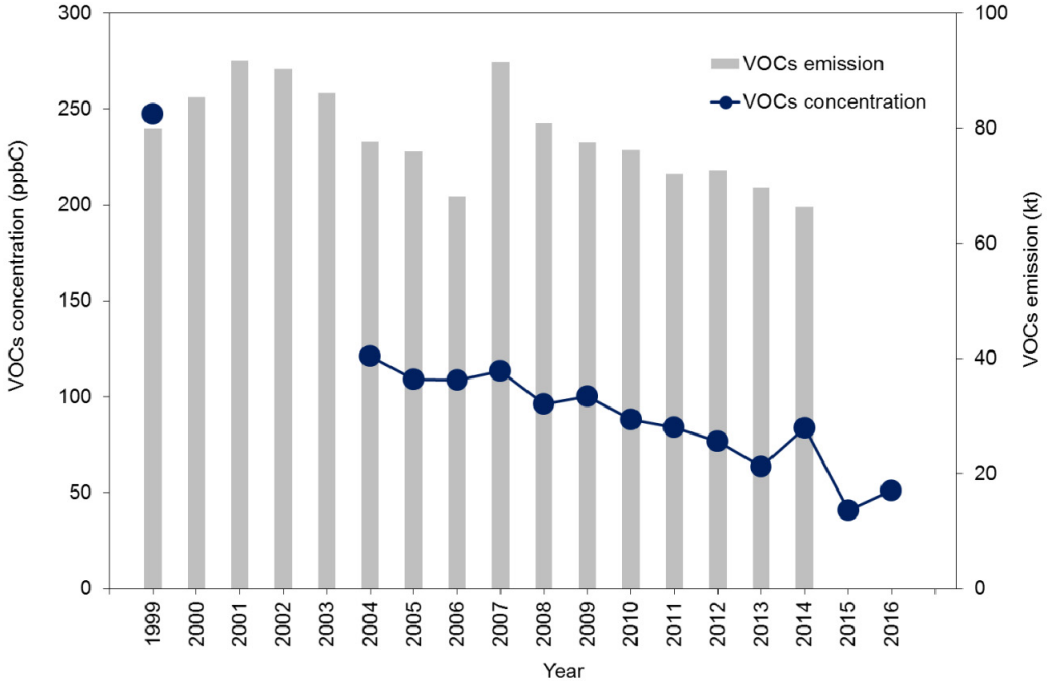

Fig. 10. Trend of the annual average concentration of total VOCs, the sum of the 55 compounds measured at Bulgwang, Seoul and emission amount of VOCs in Seoul. QA/QC protocols proposed by Shin *et al.* (2011) were applied to the raw data. Data are from NIER except the data between 1998 and 1999 (shown as 1999) which are from Na and Kim (2001).

Table 3. Comparison of the major sources of VOCs in Seoul between emission inventory data and receptor modeling results.

Fraction (%)	Vehicular exhaust	Solvent usage	Others
2009 (Emission inventory)	24 (gasoline evaporation not included)	69	7
PMF (2 sites) (2009–2010)	44–48 (gasoline evaporation included)	31–37	-
2001 (Emission inventory)	28.8 (gasoline evaporation not included)	65.0	6.2
CMB (1998–2000)	52 (gasoline evaporation included)	26	22

Emission inventory data in 2001 and the CMB result are from Kim (2009), and emission inventory data in 2009 are calculated from the data from MOE (2011) and the PMF result is from SI (2012).

for their measurement between 2004 and 2008. They identified three factors, gasoline vapor, vehicular exhaust, and solvent evaporation with decreasing variance. Thus, all research results have pointing the inconsistency between the emission inventory (solvent usage being the dominant source) and the measurement based analyses (vehicle related emissions being the dominant source). Further research is warranted to resolve this problem.

Na and Kim (2007) estimated the ozone forming potential (OFP) of the measured VOCs by weighting with values for the maximum incremental reactivity (MIR) (Carter, 1994). They reported that the OFP from vehicular exhaust was the largest (52%) followed by solvent usage (29%) though solvent usage contributed slightly more in per mass basis than vehicular exhaust. Based on the measurement of aromatics between November 1997 and July 1999, Na *et al.* (2005) estimated source contribution to them by factor analyses and estimated the OFP of each source. On an average, about 60% of the measured aromatics were estimated from vehicular exhaust remaining from evaporative emissions including solvent usage and gasoline evaporation. Xylenes were the most dominant contributor to the ozone formation followed by toluene though the concentration of toluene was higher than xylenes. Shin *et al.* (2013a) also estimated the OFP using the photochemical ozone creation potential (POCP) values by Derwent *et al.* (2007) and secondary organic aerosols (SOA) formation potential using the SOA yield values. Toluene had the highest contribution to ozone formation in Seoul, accounting for about 28% of the total ozone formation, due to its high concentration in Seoul and high photochemical reactivity followed by ethylene (10.9%) and *n*-butane (8.1%). For the SOA formation, aromatics played dominant role with toluene contributed 55% of the SOA production in Seoul followed by benzene (16.2%), ethylbenzene (6.7%), and *m*-/*p*-xylenes (5.2%). The total SOA formation potential of VOCs ranged from 2.5 to 3.5 μg m^{-3}. From these results, Shin *et al.* (2013a) proposed the control strategy of VOCs for Seoul to reduce both ozone and SOA should be to control both on solvent usage and traffic related emissions. They further argued that the control of emissions of high carbon alkanes, which are emitted from meat cooking and

gasoline powered motor vehicles with high emission rates, should also be considered due to their relatively high SOA formation potentials (Shin *et al.*, 2013a).

Ozone

Since most ozone in urban areas is generated in the troposphere by photochemical reactions, the fate of ozone is closely related with that of fine particles a majority of which is also generated by photochemical reactions in the air. However, long-term ozone trends were often decoupled with those of aerosol in many urban areas in northeast Asia. Especially in China and Korea, levels of both PM_{10} and $PM_{2.5}$ have gone down in accordance with reduced coal use and strict emission controls in recent years, but ozone continues to rise at alarming levels (Ghim *et al.*, 2015; Shin *et al.*, 2015; Lang *et al.*, 2017).

Shin *et al.* (2012) studied the meteorologically adjusted long-term variations of the surface ozone concentrations over 6 major cities in Korea including Seoul using the low-pass Kolmogorov and Zurbenko (KZ) filter, multiple linear regressions for the data between 2001 and 2008. During the period, the ozone concentration in Seoul has increased by 3.3% y^{-1}. Interestingly, that at Incheon, another large city in the SMA with a population of ~2 million and about 40 km from Seoul, has decreased (–1.1% y^{-1}). It suggested that the local emission characteristics might be important to the ozone level in the city. They tried to separate the variation of ozone induced by the change of precursor emissions from the long-term variation of ozone by using the OZone Isopleth Plotting package for Research-oriented version (OZIPR) based on the official emission inventory of Korea, the CAPSS. They claimed that although both the effects of precursor emission and background level change have affected the increase of the surface ozone concentration from 2001 to 2008, the background effect is about 2.5 times higher than the precursor emissions change on the surface ozone concentration over Seoul (Shin *et al.*, 2012).

In recent studies, Kim *et al.* (2018) have also confirmed rather consistent 0.65 ppbv increase of ozone per year in SMA from 2000 to 2016, while yearly average of daily maximum 8-hour O_3 in summer season had a much higher rate of 1.2 ppbv per year (Fig. 11). They explained the increase of O_3 mainly by reductions of NO emission, but also indicated that the rising background O_3 in northeast Asia was likely because of the observed O_3 increase. The possibility of enhanced background (regional) ozone in the SMA was previously recognized in the spatiotemporal studies of ozone with land-use types in Korea (Yoo *et al.*, 2015). Recent increases of ozone and its precursors in China could affect regional ozone levels in East Asia (Seo *et al.*, 2014). Previous studies have shown that ozone transport from China with continental outflow is a major source of ozone in Japan and South Korea (Tanimoto *et al.*, 2005; Nagashima *et al.*, 2010). A potential source contribution function was also used to determine the probable regional and local sources of ozone in Seoul (Vellingiri *et al.*, 2016). This study suggested that both regional and urban emission sources were comparable pathways to the recent increase in ozone at the study site.

Several studies have been carried out using the OZIPR to Seoul and the SMA for different periods. For example, Park and Kim (2002) applied the OZIPR for the three cases, August and September 1998 and June 1999 with the CAPSS. They found that during those periods, Seoul was in the VOC limited condition. Jin *et al.* (2012) simulated for Seoul (4 sub-areas), Incheon (4 sub-areas), and 24 cities in the SMA for the period June 2000. They found that Seoul and Incheon and 12 cities in the SMA were in the VOC

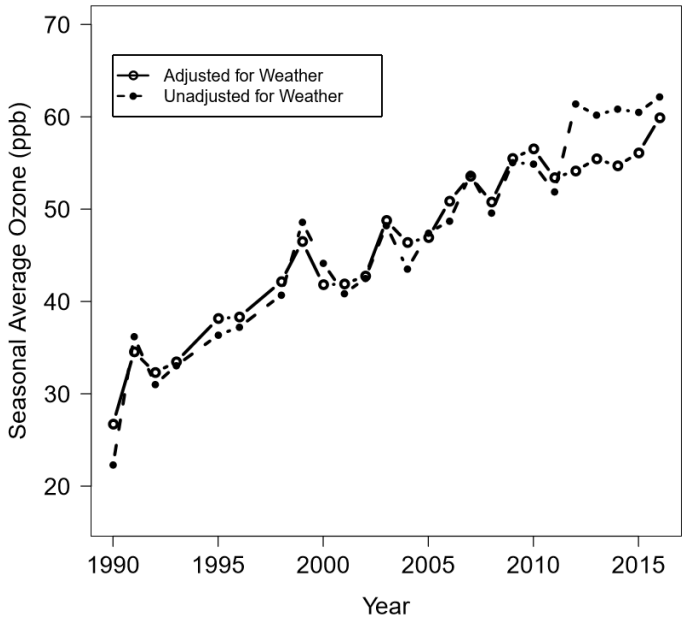

Fig. 11. Average of the daily maximum 8-hour O_3 concentrations from 1990 to 2016 in the SMA. The dotted line with closed circle shows the trend in observed O_3 concentrations in the SMA, while the dotted line with open circle shows the underlying O_3 trend at those sites after removing the effects of weather (Kim *et al.*, 2018).

limited condition while the other 12 cities were between the VOC limited and NO_x limited condition. They also reported that the simulated maximum ozone concentrations for all cities were lower than the observed ones.

Uncertainties in ozone-VOCs-NO_x sensitivity in semirural SMA were extensively discussed with respect to missing OH reactivity related with ozone and secondary aerosol formation (Kim *et al.*, 2015). They suggested that oxygenated VOCs (OVOCs) and HONO, which were rarely measured in the SMA but are likely significant sources and sinks of radicals, might play a major role in additional ozone and secondary aerosol formation. These unconstrained OVOCs could cause higher-than-expected OH loss rates and rapid ozone and secondary organic aerosol formation in semirural areas of the SMA.

Visibility

Visibility is an excellent indicator of air quality because its impairment can be easily recognized even with the naked eyes of lay people. In Fig. 12, the trend of the annual mean visibility in Seoul is shown. During last three decades, the mean visibility has increased a couple of kilometers.

Ghim *et al.* (2005) studied the trend of visibility in Korea in general and Seoul in particular between 1980 and 2000. They found that the yearly mean visibility decreased nationwide during the 1980s and 1990s while it increased in Seoul in the 1990s as also shown in Fig. 12. They also suggested that the visibility in winter morning was lower than the summer afternoon can be attributable to higher aerosol concentration associated with lower mixing height and higher ambient relative humidity (Ghim *et al.*, 2005).

Since visibility is closely related with the characteristics of aerosols (size, chemical composition, and concentration), an indicator of air quality, it can be seen that air quality in Korea rapidly degraded in the 1980s and 1990s. Also, it can be seen that air quality in Seoul was continuously improved in the 1990s. This trend in Seoul is consistent with the decreasing trend of PM_{10} shown in Fig. 4.

REMAINING PROBLEMS AND FURTHER RESEARCH DIRECTIONS

Despite of progressive improvement of air quality in the SMA, our scientific understandings are still far too incomplete and current emission reduction plans are often questionable to attain desired air quality in near future. Large discrepancies and uncertainties still exist mainly in estimating precursor emissions, contributions from regional and local sources, and characteristic formations of ozone and secondary aerosol in the SMA.

Uncertainties in National Emission Inventories

Although emission inventory, CAPSS has been improved extensively in recent years, emissions of major precursor gases remain highly uncertain in the SMA. Mostly, CAPSS still underestimates current and historical levels of major precursors, such as NO_x and VOCs, which is mainly due to unidentified emission sources. It is highly likely that CAPSS has missed unknown significant NO_x emission sources such as diesel vehicles. In addition to missing local NO_x sources, we should not exclude the possibility of regionally transported contributions of NO_2 or longer NO_x photochemical chain-lengths that could extend the chemical lifetime of NO_2. The latter may explain the increasing

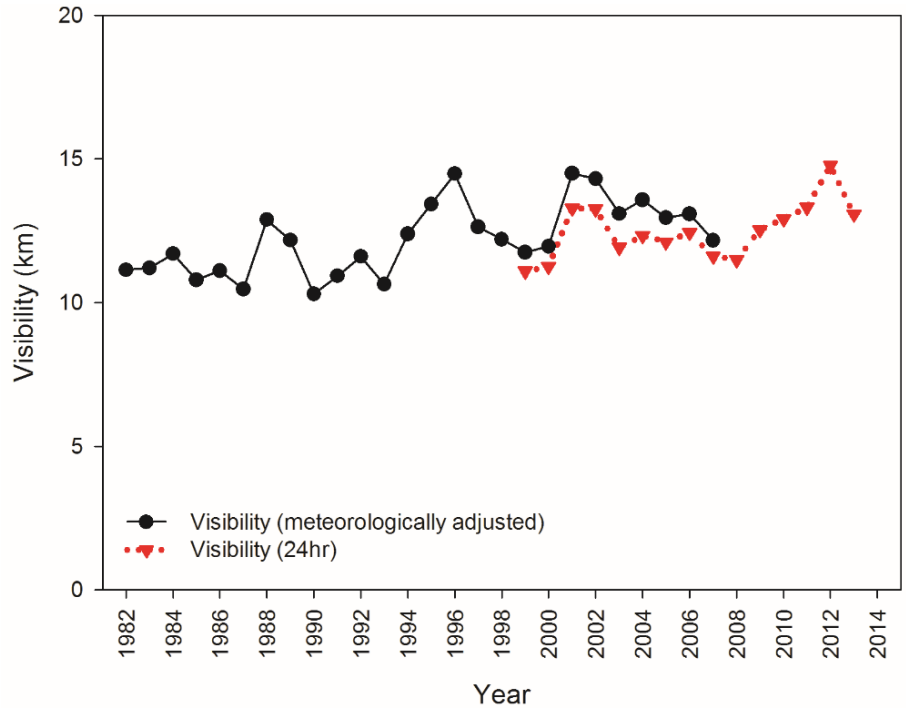

Fig. 12. Trend of the visibility in Seoul between 1982 and 2013. Meteorologically adjusted visibility is estimated by removing the data of precipitation day. Data are from the Korea Meteorological Administration.

trend of secondary photochemical pollutants such as O_3 because they are direct by-products of the NO_x chain cycle (Lee *et al.*, 2015).

It is also important to improve anthropogenic and natural biogenic VOC (BVOC) emissions which is closely connected to the production of ozone and secondary aerosol. Current anthropogenic VOCs and CO emissions are clearly underestimated to explain their ambient levels and BVOCs have not been clear to constrain reactive VOCs budgets in the SMA (Lee *et al.*, 2012). VOC emission reduction has been much slower than that of NO_x in Korea. It is well known that photochemical air pollution can be worsen under increasing ratios of VOCs/NO_x with decreasing concentrations of NO_x in most metropolitan areas (Seinfeld and Pandis, 2016). Our understanding of recent increase of O_3 and organic components in secondary aerosol in SMA are largely hindered by unconstrained VOC and NO_x emission inventories.

Uncertainties in Regional Contributions on Air Quality

Air quality in the SMA depends mostly on local sources such as mobile, domestic and industrial emissions. However, many studies indicated that significant portions of secondary aerosol in the SMA were clearly influenced by regional sources other than local emissions. In transport model analysis with long-term monitoring data, overall regional source contributions accounted for 8–21% for SO_2 and 13–20% of OC and BC (Jeong *et al.*, 2011; Jeong *et al.*, 2013) in the SMA. It is well known that fast moving continental outflows play an important role sporadically with much intensive levels of aerosol to the SMA (Park *et al.*, 2011). Especially, this impact of continental outflows from Asian continent has been apparent over northeast Asia and has been greater in recent years (Lee *et al.*, 2013; Yang *et al.*, 2013). Nevertheless, trans-boundary contributions for both aerosol and ozone in the SMA using chemical transport models have not been fully validated due to uncertainties in emission and limited comprehensive observations.

Another complication by regional contributions in the SMA is rapid chemical transformation during the transport process from source regions. Cayetano *et al.* (2011) found that the chemical compositions in aerosol particles were changed during their passage over the Yellow Sea from continent source regions. Although key chemical components of aerosols were mainly retained to those of source regions, distinct enrichment of acidic components was observed during transport. Also, when sufficient precursors were available in a fast-moving cold and dry air mass in a frontal system from the Asian continent, new particle formations were often witnessed in the Yellow Sea (Kim *et al.*, 2013c). To resolve both accurate short-term and long-term regional influences to the SMA, comprehensive and integrated studies with model, observations, improved emission sources are highly critical.

One important regional transport of air pollutants to the SMA is North Korea which is just north of the SMA. Lee and Kim (2007) and Kim *et al.* (2013a) reported that the polycyclic aromatic hydrocarbons (PAHs) observed in Seoul might be significantly impacted by North Korea biomass burning activities.

Unresolved Chemistry in Ozone and Secondary Aerosol Production

PAN (Peroxy Acetyl Nitrate), which is a photochemical indicator species and very sensitive to reactive VOCs and NO_x, increased about twice in the Yellow Sea since late 1990s and average PAN/O_3 ratios in the SMA was significantly higher than those observed in European and North American cities (Lee *et al.*, 2008; Lee *et al.*, 2012). These studies showed that levels and compositions of VOCs in the SMA are particularly favorable for rapid production of this photochemical species. Despite the importance of VOCs in PAN, ozone, and $PM_{2.5}$ chemistry, its detailed information in the SMA has been limited to a few studies (Na *et al.*, 2003, 2004; Shin *et al.*, 2013a, b). In addition to anthropogenic VOCs, Kim *et al.* (2013c) determined that BVOC photochemistry could play a significant role in ozone formation in the suburban region of the SMA. In another study, they found that observed total OH reactivity in the SMA was well matched by the observed trace gas dataset, but observed total OH reactivity in the suburban forest area could not be largely accounted for (~70%) by the extensive trace gas measurements (Kim *et al.*, 2016). They claimed that the underestimation of BVOC emissions was the main cause for this missing OH reactivity in the semirural SMA. The detailed VOC compositions including biogenic and oxidized VOCs are critical to understand how productions of ozone and secondary aerosols are particularly high in the SMA.

Another potential source of ozone and aerosol production is the pre-existing aerosol surface. With recent studies, more evidences have been shown that heterogeneous reactions on aerosol surface enhanced oxidant production and aerosol growth (Palacios *et al.*, 2016; Lim *et al.*, 2017; Yu *et al.*, 2017). This additional source of oxidants and aerosol is very probable in SMA where high aerosol concentration sustains.

Heterogeneous sources of NO_3 and Cl radical are particularly important during the night and the early morning of consequent day. Brown *et al.* (2017) illustrated that the nighttime chemistry of nitrogen oxides and O_3 were rapid in Seoul with high nitrate radical production rates, $P(NO_3)$, averaged 3–4 ppbv h^{-1} in late afternoon and early evening, much greater than contemporary data from Los Angeles, a comparable U. S. megacity. It is reasonable to consider that these nighttime radicals may play significant roles in the production of oxidized hydrocarbons, PAN, ozone and secondary aerosols in the SMA. However, systematic and concurrent observations with other species were not made in the SMA.

Future Researches

Amount and natures of emissions, and related chemistries change over the times. Performance of air quality policy should be evaluated according to proper timeline with taking account of scientifically sound perspectives and evidences. However, systematic and scientific accountability on air quality measures has been very scarce in the SMA. It is not

surprising air quality in the SMA has been staggering despite of growing emission reduction efforts, especially in recent years (Kim *et al.*, 2017). Current discoursed uncertainties and unknowns related with the SMA air quality are major sources of difficulties in implementing confident control strategies and emission reduction plans onward.

The recent Megacity Air Pollution Studies (MAPS)-Seoul program was the first integrated scientific effort in the SMA to assess critical questions in the extensive context of emission, chemical transformation and transport. Two intensive field campaigns utilized ensemble of chemical transport models, newly adapted emission inventories, comprehensive chemical observation, and satellite monitoring (Lee *et al.*, 2015). Its outcomes started to reveal scientific clues for many unresolved questions as they are published in companion papers in this special issue. However, as this program covered only a limited time of year (May–June), the continuous and successive efforts for detailed sciences and researches should be followed in the imminent future.

Many studies implied that aerosol productions could be clearly enhanced in wintertime (Sun *et al.*, 2016) and, nighttime VOCs oxidation by NO_3 was comparable to that from daytime OH oxidation pathways (Sobanski *et al.*, 2017). Although previous studies of radical budgets in the SMA indicated the importance of nighttime chemistries, quantitative assessments on these issues were not fully made until this moment (Brown *et al.*, 2017; Kim *et al.*, 2016). In future, air quality research should have special attention on nighttime and heterogeneous chemistry, especially during the wintertime on coastal SMA where large sources of NO_x and Cl are available.

With thorough and extensive revision of emission inventories in recent studies, their uncertainties have been reduced in many species in the SMA (Li *et al.*, 2017). However, further improvements should be made, especially for precursor species, such as VOCs, NO_x, and NH_3 whose uncertainties are still larger than 50% (Crawford *et al.*, 2017). It is well known that current bottom-up approach in CAPSS emission model has the inherited errors accumulated along subsequent processes and steps. In contrast, top-down emission approach mainly with inverse model is proven to have greater accuracy in representing observed values even in complex situations (Thompson *et al.*, 2016). However, it is also true that top-down approach may result in significant bias with allocating unrealistic factors to local and individual activities. Thus, future emission studies are required to use both approaches for complementary purpose and cross-checking.

The satellite and ground remote- sensing techniques are invaluable parts of integrated air quality researches. They provide concurrent and wide spatial coverage of aerosol and major trace gases. Although they have been heavily used to analyze the regional variations of interest species, increased number of studies have been made to yield air quality analysis in urban and its surrounding vicinity (Tulloch and Li, 2004). Particularly, recent geostationary satellite sensors, such as GOCI (Geostationary Ocean Color Imager) and HIMAWARI, provide unprecedent high resolution temporal and spatial data for urban air quality, but limited to aerosol optical depth (Lee *et al.*, 2010; Yumimoto *et al.*, 2016). GEMS (Geostationary Environmental Monitoring Spectrometer) will provide a new generation of satellite sensor for aerosol and major gases, which is expected to launch within a year (Kim, 2012). With these newly available research tools, our understanding in air quality forecasting, emission validation, and long-range transport of air pollutions will be greatly improved.

SUMMARY

Based on the available data, the air quality in Seoul has improved during the last four decades due to government policy measures on air pollution. The concentrations of primary air pollutants decreased drastically during the 1980s and 1990s due to the strict regulation of fuels and emission standards. Even with the strict regulations, however, the number of emission sources has increased along with the standard of living; as a result, the overall levels of air pollutants have not decreased since the early 2000s. To improve the air quality in the SMA, a special act was enacted, and significant funding was allocated, mostly for the control of emissions from diesel vehicles. The mass concentrations of $PM_{2.5}$ and PM_{10} and the concentrations of OC and EC in the $PM_{2.5}$ have shown a general decreasing trend since 2003.

However, the concentrations of inorganic ions in $PM_{2.5}$, such as sulfate, nitrate, and ammonium, have not exhibited a decreasing trend, and it has been recognized that the major contributors to these ionic species have not been clearly identified. Furthermore, the visibility in Seoul has not improved much.

These observations clearly indicate that our understanding of air quality improvement in Seoul has been limited to the early stages of information processing in using simplified emission inventories and relying on apparent chemical behaviors. The initial step of effectively implementing air quality measures in the SMA consists of identifying the unknowns and reducing uncertainties in our understanding of air quality, which requires addressing all aspects of emission sources, transport, chemical transformation, and sinks.

The high research priority for policy implementation is identifying the missing emission sources of ozone and aerosol in the SMA, especially for VOCs. The uncertainties in emission inventories for VOCs are the highest among the precursor species, and the reduction of VOC emissions has been very slow by comparison. Although recent studies have indicated that nighttime and wintertime radical chemistries associated with VOCs and NO_y species may play significant roles in ozone and aerosol production in the SMA, clear and quantitative evidence is still limited. Continuous research with integrated sciences utilizing all available resources and platforms, such as MAPS-Seoul, is required to resolve complex urban air quality issues. In addition to these current efforts, upcoming satellite platforms with extensive temporal and spatial coverage over Asia, including GEMS, will greatly enhance our capacity to understand urban and regional air quality.

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