The Effects of Precursor Emission and Background Concentration Changes on the Surface Ozone Concentration over Korea

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ABSTRACT

The meteorologically adjusted long term variations of the surface ozone concentrations over 6 major cities in Korea were determined by using the low-pass Kolmogorov and Zurbenko (KZ)-filter and multiple linear regressions. In addition, by using the OZone Isopleth Plotting package for Research-oriented version (OZIPR), the variation of ozone induced by the change of precursor emissions was separated from the long term variation of ozone over Seoul, thus the relative contribution of the background ozone concentration was deduced. Although both the effects of precursor emission and background level change have increased the surface ozone concentration from 2001 to 2008, it is inferred that the background effect is about 2.5 times higher than the precursor emissions change on the surface ozone concentration over Seoul. The relative importance of the background effect might be increasing as the emissions in Northeast Asia increase, while the effect of precursor emission could be reduced by continuous implementation of emission control strategies in Seoul.

Keywords: Ozone; Low-pass KZ-filter; Emission effect; OZIPR; Long range transport.

INTRODUCTION

Atmospheric surface ozone concentration or mixing ratio has increased continuously since air quality monitoring network was established in 1989 in Korea. Also the number of days of ozone warning which is issued at the 1-h ozone concentration above 0.120 ppm has increased up to 32 days in 2008 from 17 days in 2000 (KMOE, 2009a). High ozone concentration can have adverse effects on human health, natural environment, and agricultural yields, and a significant impact on air quality and climate change (Berntsen *et al.*, 1996; Chameides *et al.*, 1999; Mauzerall *et al.*, 2005).

To reduce the atmospheric surface ozone concentration, several measures including reduction of the allowable emission rate for volatile organic compounds (VOCs) in various emitting facilities and installation of emission reduction equipments to vehicles have been enforced since 1999 (KMOE, 1999). Korean Ministry of Environment (KMOE) has established the Photochemical Assessment Monitoring Stations (PAMS) to monitor the concentration of ozone precursors, VOCs and nitrogen oxides (NO_x)

responding to high atmospheric ozone phenomenon since 2002.

It is important to distinguish the effect of precursor emission and meteorological factors on the atmospheric surface ozone concentration because ozone is not a primary pollutant, but a secondary pollutant which is produced by photochemical reactions of precursor species under certain meteorological conditions (Seinfeld and Pandis, 2006). In other words, atmospheric surface ozone concentration could be changed with changing meteorological factors only, without change of precursor emission amounts. Thus, to assess the effectiveness of ozone control measures, quantification of the variation of ozone concentration caused by meteorological factors has to be carried out in advance.

Another variable that affect the ozone concentration in Korea is the regional background concentration of ozone over the Asian continent. According to Tanimoto *et al.* (2005), the most intensive influence of outflow from the Asian continent was centered at 30–40°N, where the Korean Peninsula is located. The "regional background" is defined as the variable caused by the change of long term emission trend of ozone precursors in the Asian continent and the substantial transport of photochemically produced ozone to the downwind area of the Asian continent including Korean Peninsula.

The high ozone phenomenon is no longer confined to Korea. The background ozone concentration over Northeast Asia including Korea is higher than western Europe and the

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USA by 5–10 and 10–30 ppb, respectively (Fiore *et al.*, 2001; Pochanart *et al.*, 2004). In addition, the background ozone concentration has increased continuously (Vingarzan, 2004). Thus, the background effect should be also separated to estimate the effect of ozone control strategy in Korea. Kim *et al.* (2003) proposed that it is needed to consider the contribution of increasing background ozone concentration in Northeast Asia for ozone trend analysis in Korea because ozone increasing rates in Busan and Seoul were higher than those in USA.

From the early 1990s, the low-pass Kolmogorov and Zurbenko (KZ)-filter method, a low pass time-series filtering method, has been widely used to detect meteorologically adjusted long term variation of atmospheric pollutants, such as ozone and particulate matter (Rao and Zurbenko, 1994; Milanchus *et al.*, 1998; Ibarra-Berastegi *et al.*, 2001; Lu and Chang, 2005; Wise and Comrie, 2005; Lee *et al.*, 2008). With the low-pass KZ-filter method, one can decompose pollutant concentration and meteorological factors to several time series components with different temporal level. Thereafter, the multiple regression is usually applied to the filtered pollutant concentration (dependent variable) and meteorological factors resolved by the low-pass KZ-filter method (independent variables). Then, the meteorologically adjusted pollutant concentration, which is generally assumed to be the same with the filtered residual of regression by large filter width (1.7 year), is produced (Rao and Zurbenko, 1994; Milanchus *et al.*, 1998; Ibarra-Berastegi *et al.*, 2001; Lu and Chang, 2005; Wise and Comrie, 2005).

Even though long term variation decomposed by the low-pass KZ-filter method and multiple regression is generally assumed to be the only signal caused by emission change, the long term variation in Korea is thought to be divided to two sub-components; the regional background variation (regional background effect, B) and the domestic precursor emission induced variation (emission effect, E).

The OZone Isopleth Plotting package for Research-oriented version (OZIPR) is a trajectory type, air quality simulation model based on the US EPA's OZone Isopleth Plotting Program (OZIPP) model, utilizing complex chemical mechanisms, emissions, and various meteorological factors (Gery *et al.*, 1990). The OZIPR is generally used to simulate the ozone episode occurred in urban area and thus, to support the policy makers deciding effective emission reduction strategy for improving air quality (Park and Kim, 2002; Hong *et al.*, 2005; Do *et al.*, 2007; Orlando *et al.*, 2010).

In this study, the meteorologically adjusted atmospheric surface ozone concentrations (long term variation, L) of 6 major cities in Korea; Seoul, Incheon, Busan, Daegu, Daejeon, and Gwangju were determined by separating meteorological effects using the low-pass KZ-filter and multiple linear regression method. Then, using the OZIPR, the variation of the 1-h maximum ozone concentration for Seoul due to the changes of domestic ozone precursor emission (E) was simulated. By comparing the relative variation of the meteorologically adjusted ozone (L) and emission induced ozone concentration (E), the effect of the regional background ozone concentration (B) was deduced. Also, the component of episodic long range transport of ozone, which is assumed

to be resolved in the decomposed short term variation in the low-pass KZ-filter method, was further studied by using a simple graphical wind-sector grouping method.

The result of this study can be applied to assess the effectiveness of the proposed emission control measures to reduce the atmospheric surface ozone concentration in Seoul, and to fill the gaps in the ozone data in Northeast Asia.

MATERIALS AND METHODS

Data Acquisition

The ozone data analyzed in this study have been measured at the nation-wide air quality monitoring stations by the UV photometric method since 1989. In this study, the data of major 6 cities (27 sites in Seoul, 15 sites in Incheon, 12 sites in Busan, 11 sites in Daegu, 7 sites in Daejeon, and 7 sites in Gwangju) were used. The statistical summary of the ozone data measured in major 6 cities is given in Table 1. The locations of each air quality monitoring sites in 6 major cities are shown in Fig. 1. The hourly ozone concentrations of each monitoring site which were collected and managed by the National Air Quality Data Center (NAQDC), National Institute of Environmental Research (NIER), were calculated to produce the daily 1-h maximum concentration by cities for the time period from 2001 to 2008. Every 1-h ozone concentration of each station in each city was averaged to create 1-h ozone concentration for each city and then the daily maximum was determined by city. The time scales used in ozone trend analysis is various from 5 min to daily summaries, but the daily 1-h maximum concentration is the most widely used in the previous studies (Thompson *et al.*, 2001). Measurement data of the ozone precursors, hourly NO, NO_2, and VOCs, which were used to make input data for OZIPR running, were also provided from NAQDC, NIER.

Meteorological data at the Regional Meteorological Observatory for each city were obtained from the Korea Meteorological Administration for the same time period and resolution. Meteorological factors of interest were daily mean ambient temperature (TA), wind speed (WS), relative humidity (RH) and solar radiation (SR) which have been mostly revealed as the common meteorological factors relevant with ozone formation in several previous studies, and daily mean atmospheric pressure (PA) and dew point temperature (DT) as the additional variables (Thompson *et al.*, 2001; Wise and Comrie, 2005). The statistical summary of the meteorological parameters are given in Table 2.

The annual emission data of VOCs, NO_x, and CO for 6 cities were obtained from the Clean Air Policy Support System (CAPSS), which is an affiliated organization to manage the national emission inventory under NIER. Because the method for calculating the amount of emission has been changed since 2007, the annual emission data only from 2001 to 2006 were used for consistency.

Decomposition of Ozone Concentration and Removing Meteorological Effects

The time series of the logarithm of the daily 1-h maximum ozone concentration can be decomposed into three components; short term variation, seasonal variation,

Table 1. Statistical summary of the 1-h ozone concentration measured in 6 major cities in Korea between 2001 and 2008.

(unit: ppb)

		2001	2002	2003	2004	2005	2006	2007	2008
Seoul	Mean	15	14	13	14	17	18	18	19
	S.D.	13	13	13	13	15	15	16	16
	Max.	98	122	85	96	135	106	143	122
	Min	2	1	1	1	1	1	1	1
Incheon	Mean	19	19	20	20	22	21	22	23
	S.D.	13	13	15	14	14	13	15	16
	Max.	103	103	101	97	108	88	116	111
	Min	1	2	2	2	2	2	2	2
Busan	Mean	25	24	24	25	23	24	24	26
	S.D.	13	13	14	14	13	13	13	14
	Max.	94	86	100	86	91	107	86	95
	Min	4	3	2	3	2	2	2	2
Daegu	Mean	19	18	21	22	21	20	21	24
	S.D.	15	13	17	17	16	16	17	18
	Max.	112	88	116	139	93	95	99	106
	Min	2	2	1	3	2	1	1	1
Daejeon	Mean	21	19	19	20	21	18	17	23
	S.D.	16	14	16	17	18	14	13	18
	Max.	108	86	94	114	107	82	82	98
	Min	2	2	2	1	1	1	2	2
Gwangju	Mean	19	16	18	22	22	22	23	23
	S.D.	13	12	12	15	16	17	16	15
	Max.	87	87	84	118	98	104	105	80
	Min	1	0	2	1	1	1	1	2

S.D.: Standard Deviation.

and long term variation by the low-pass KZ-filter produced by a moving average of a time length (*m*) and iteration (*p*) (Ibarra-Berastegi *et al.*, 2001). The precise methodology of the low-pass KZ-filter applied to ozone filtering can be found elsewhere (Ibarra-Berastegi *et al.*, 2001). In this study, effective filter width (*Np*), which was the period of the smallest frequency removed, was decided to be 50 days for removing periodicities (*m* = 29, *p* = 3) smaller than seasonality according to (1).

$$m \times p^{1/2} \leq Np \tag{1}$$

After applying the $KZ_{m=29,\ p=3}$ filter, the observed ozone concentration could be expressed by Eq. (2).

$$O_3(t) = O_{3KZ}(t) + O_{3ST}(t) \tag{2}$$

where, $O_3(t)$ is the logarithm of the daily 1-h maximum ozone concentration. And $O_{3KZ}(t)$ and $O_{3ST}(t)$ are the summation of the seasonal and long term variation, and short term variation of ozone, respectively.

The same low-pass KZ-filter method was applied to the meteorological factors as the first step to remove the meteorological effects from $O_{3KZ}(t)$, thus the results of $KZ_{m=29,\ p=3}$ filter for i^{th} meteorological factor $(MET_{KZ}(t))_i$ were produced.

The next step was to perform the multiple linear regression between $O_{3KZ}(t)$ (independent variable) and $MET_{KZ}(t)_i$ (dependent variables) as shown in Eq. (3).

$$O_{3KZ}(t) = a_0 + \sum a_i MET_{KZ}(t)_I + \varepsilon(t) \tag{3}$$

where, a_i is the regression coefficient and $\varepsilon(t)$ is the residual of the regression. The residual $\varepsilon(t)$ is the component which consists of long term variation of ozone $(\varepsilon_{KZm=365,p=3}(t))$ and seasonal variation related with other meteorological factors $(\delta(t))$ not included in the multiple linear regression (Eq. (4)).

$$\varepsilon(t) = \varepsilon_{KZm=365,p=3}(t) + \delta(t) \tag{4}$$

Then, seasonal variation of ozone $(O_{3SEASON}(t))$ was defined by combination of the seasonal components of Eq. (3) and Eq. (4).

$$O_{3SEASON}(t) = a_0 + \sum a_i MET_{KZ}(t)_i + \delta(t) \tag{5}$$

Thus, the logarithm of the daily 1-h maximum ozone concentration was decomposed into three different components as shown in Eq. (6):

$$O_3(t) = \varepsilon_{KZm=365,p=3}(t) + O_{3SEASON}(t) + O_{3ST}(t) \tag{6}$$

Input Data for OZIPR

In this study, OZIPR, a photochemical box model, was used to simulate the photochemical ozone formation over Seoul. OZIPR model was run with annual precursor emission changes with fixed meteorological factors and initial precursor concentration. The result was compared with meteorologically
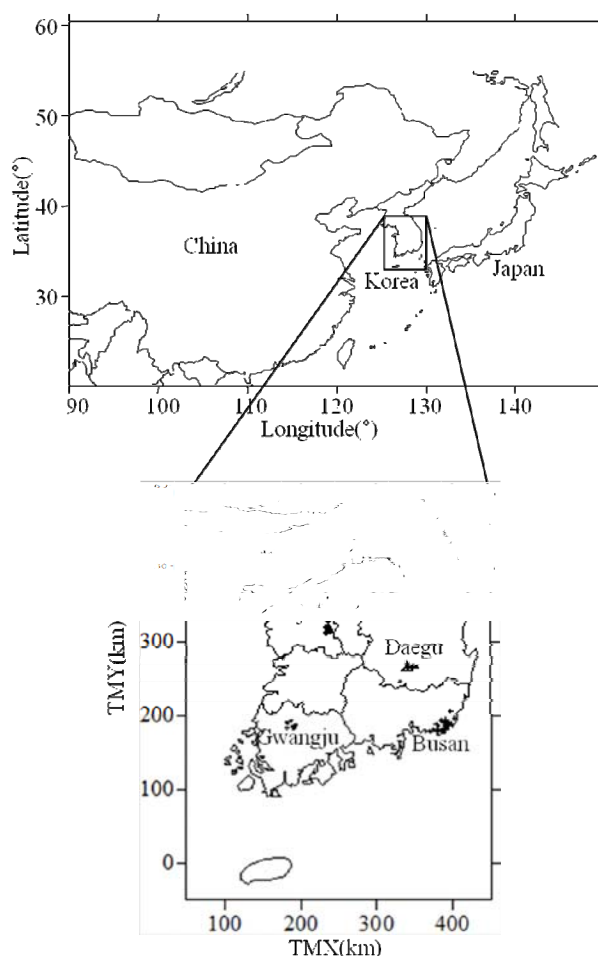
Fig. 1. Ozone monitoring stations in six major cities.

Table 2. The mean and standard deviation values of the meteorological parameters in 6 major cities in Korea between 2001 and 2008.

City	Temperature (°C)	Wind speed (m/s)	Relative humidity (%)	Solar radiation	Pressure (hPa)	Dew point temperature (°C)
Seoul	12.9 ± 10.1	2.2 ± 0.8	61.6 ± 14.7	12.1 ± 6.6	1016.1 ± 8.1	5.1 ± 11.6
Incheon	12.8 ± 9.6	2.6 ± 1.2	67.0 ± 14.4	12.7 ± 6.8	1016.0 ± 8.0	6.4 ± 11.2
Busan	14.8 ± 7.9	3.3 ± 1.2	62.6 ± 18.4	14.0 ± 6.9	1015.5 ± 7.1	7.1 ± 11.4
Daegu	14.6 ± 9.3	2.3 ± 0.9	58.0 ± 16.3	13.5 ± 6.3	1015.9 ± 7.6	5.5 ± 11.6
Daejeon	13.2 ± 9.7	1.9 ± 0.9	65.7 ± 13.6	13.7 ± 6.7	1016.2 ± 8.0	6.2 ± 10.9
Gwangju	14.1 ± 9.2	2.0 ± 0.8	66.7 ± 12.9	14.2 ± 6.8	1016.2 ± 7.9	7.5 ± 10.3

adjusted ozone long term variation, which is assumed to be the function of emission change and the variation of the regional background concentration.

To run the OZIPR, information on various variables; chemical mechanism, initial concentrations of the chemical species related with ozone formation, VOCs speciation ratio, NO_2/NO_x ratio, the transport concentrations of ozone from upper layer and surface layer, deposition rate profiles, meteorological conditions, and temporally varying emission rate were needed. The chemical mechanism used in this study was Regional Acid Deposition Model (RADM2). The default values were used for deposition rate profiles and the mixing height. To assess the emission induced

change of ozone concentration, other parameters except for emission rate were fixed. The initial concentrations of VOCs, NO_x, and CO were represented by the data for the maximum ozone concentration day of Seoul in 2004 (10th August, 2004). The VOC lumping result by Hong *et al*. (2005) for VOCs speciation was used since that result was based on the Seoul measurement data. The NO_2/NO_x ratio was averaged for 9 years data from 2001 to 2009 measured in the representative Seoul monitoring site. The transports from aloft and surface were ignored since we are interested in the emission induced ozone formation. These might cause substantial uncertainty for total amount of ozone formation. However, uncertainty related with this assumption is not

considered, because relative amount of ozone produced from emission induced process, not for absolute amount of ozone, is used for further processes in this study. The atmospheric temperature was also represented by the data for the same day of initial concentration of precursors. The annual emission was converted to the temporally assigned emission rate by dividing the area of Seoul and by multiplying the temporally weighted value according to the sources provided by the US EPA (Lee, 2004). The parameters used in running the

OZIPR are presented in Table 3.

RESULTS

Characteristics of the Time Series Ozone Concentration

The logarithm of the daily 1-h maximum ozone concentration and each component decomposed to short term, seasonal, and long term variation of ozone in 6 cities are shown in Fig. 2. The episodic increase or decrease of ozone

Table 3. Input parameters for OZIPR modeling.

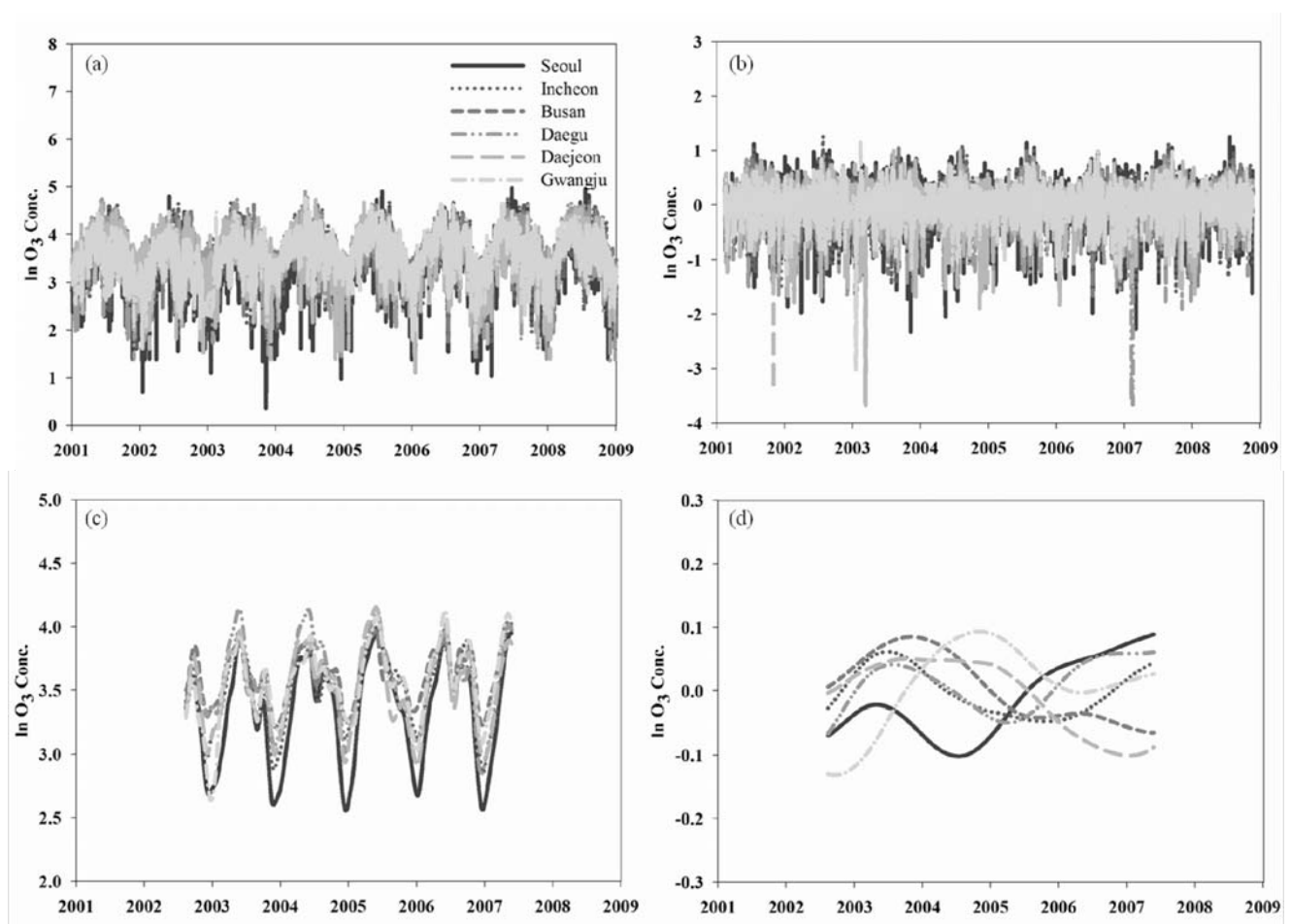|	Parameter	Value
Chemical Mechanism	RADM2	-
Initial Condition	Calculate	VOCs: 0.286 ppmC
		NO$_x$: 0.037 ppm
		CO: 0.5 ppm
Boundary Condition	Reactivity	VOCs speciation
	Ifraction	0.55
	Transport	None
	Deposition rate profiles	Default
Meteorological Condition	Dilution	Default
	Temperature	298–302 K
Emission	Emissions	Temporally varying emission

Fig. 2. Decomposed components of the ozone concentration in 6 cities: (a) Daily maximum ozone concentration, (b) short term variation, (c) seasonal variation, and (d) long term variation.

concentration occurred within 50 days interval was screened and dissolved in short term variation term. Thus, the effect of episodic long range transport was included in short term variation. The seasonal variation was similar in all cities; high in spring and low in winter due to photochemical production with the highest variation in Seoul. The annual rate of long term variation change was calculated by fitting a line to the long term variation for each city. Before fitting a trend line, logarithmic format of long term variation was converted to concentration basis by taking exponential. The slope of trend line is used to calculate the annual rate of long term variation by multiple 365 days, followed by dividing averaged long term variation concentration of each city to induce percent change. Long term variation of ozone concentrations has increased by 3.3%/y (Seoul,), 2.2%/y (Gwangju), 1.1%/y (Daegu) during the study period except for Busan (–3.3%/y), Daejeon (–3.3%/y), and Incheon (–1.1%/y). The general trend of long term ozone variation in Korea was comparable with the results of other Asian region such as China and Japan. In case of Beijing, the annual rate of change in lower troposphere below 4 km was about 2%/y during 1997–2004 (Ding *et al.*, 2008). For Tokyo, smaller increasing trend with a rate about 1%/y during the same period was reported in surface layer below 450 m (Ding *et al.*, 2008).

To quantify the effect of meteorological factors on ozone concentration, a step-wised multiple linear regressions was conducted. The total coefficient of determination (r^2), which is associated with all meteorological factors considered in this study, are shown in the left-most column in Table 4. And the increase of r^2 (Δr^2) in the top three steps of whole processes and regression coefficients in multiple linear regression are shown in right-hand columns in Table 4. The meteorological factors could explain the total variation of the ozone concentration from 66.4% (Busan) to 89.7% (Seoul). This result is consistent with the previous result of 68% (Busan) and 87% (Seoul) by Kim *et al.* (2003) in which the ozone concentration trends at Seoul and Busan for 9 years (1992–2000) were analyzed. It can be assumed that the effect of emission is the highest in Busan and the lowest in Seoul because the effect of meteorological factors on ozone concentration is the highest in Seoul and the lowest in Busan. The solar radiation was the strongest meteorological factor correlated with ozone, showing high r^2 value from 0.647 (Busan) to 0.800 (Daegu). Ambient temperature was the second major meteorological factor affecting the ozone concentration in Seoul, Incheon, and Daegu, but, relative humidity and wind speed were the second major factors in other three cities. Since the ozone formation is determined by both meteorological conditions and the levels of precursor species, the degree of the impact of each meteorological parameter varied in each city.

Table 4. Regression result of filtered ozone and meteorological variables by step-wised multiple linear regression.

O_{3KZ}	Meteorological Variables (MET_{KZ})	Coefficient		$\Delta r^{2\,a}$
		Estimated Coefficient	Std. Error (S.E)	
Seoul	Constant	1.767	0.018	
$r^2 = 0.897$	SR	0.070	0.001	0.776
S.E. = 0.132	TA	0.021	0.000	0.078
	WS	0.209	0.009	0.026
Incheon	Constant	2.458	0.019	
$r^2 = 0.821$	SR	0.049	0.001	0.740
S.E. = 0.119	TA	0.056	0.003	0.038
	DT	–0.041	0.003	0.018
Busan	Constant	–14.332	1.476	
$r^2 = 0.664$	SR	0.059	0.001	0.647
S.E. = 0.116	RH	0.006	0.000	0.015
	PA	0.017	0.001	0.002
Daegu	Constant	2.436	0.019	
$r^2 = 0.833$	SR	0.069	0.001	0.800
S.E. = 0.126	TA	0.007	0.000	0.015
	WS	0.052	0.009	0.003
Daejeon	Constant	3.688	0.072	
$r^2 = 0.816$	SR	0.049	0.002	0.784
S.E. = 0.134	RH	–0.015	0.001	0.012
	TA	0.014	0.001	0.012
Gwangju	Constant	3.022	0.033	
$r^2 = 0.785$	SR	0.086	0.001	0.697
S.E. = 0.142	WS	–0.305	0.011	0.069
	RH	–0.001	0.000	0.001

S.E.: Standard Error, SR: Solar radiation, TA: Daily mean ambient temperature, WS: Wind speed, DT: Dew point temperature, RH: Relative humidity, PA: Daily mean atmospheric pressure.
[a] The increments of r^2 (Δr^2) are calculated based on the r^2 value of former step of step-wised multiple linear regression starting with zero value.

Relative Contributions of Precursor Emission and Regional Background Level Changes

The annual total emission amount of ozone precursors, NO_x and VOCs, normalized by area of each city from year 1999 to 2006 in the 6 cities are shown in Fig. 3 (KMOE, 2009b). Although the normalized emission amounts were the highest for both NO_x and VOCs in Seoul, the normalized emission amount of precursors showed decreasing trend in Seoul till 2006 while some other cities showed increasing trend. The pattern of emission trend in each city was not directly correlated with long term variation of ozone, because ozone is the secondary pollutant produced by atmospheric photochemical reactions. As mentioned earlier, the regional background ozone concentration might be the also important factor affecting the ozone concentration over Korea.

To account for the effect of domestic emission change on the long term variation of the ozone concentration quantitatively, the OZIPR was used to simulate the photochemical ozone formation in Seoul. As mentioned before, the OZIPR model was run with the fixed meteorological factors to assure that the meteorological dependence remains constant during the modeling period.

The simulated daily maximum 1-h ozone concentration was continuously decreased from 2001 (72.6 ppb) to 2005 (51.5 ppb), and then increased up to 62.4 ppb in 2006. Even though the ozone concentration by domestic emission variation was decreased during the early 5 years (2001–2005), long term variation of ozone was increased in 2003 and 2005. If long term variation of ozone is mostly affected by domestic emission of precursors, the pattern of long term variation should be consistent with the trend of the simulated maximum ozone concentration. Thus, it suggested that there are other factor(s) contributing to long term variation of ozone in Seoul. Atmospheric surface ozone concentration contains a regional background contribution reflecting transport from outside of domestic domain, so-called regional background effect.

The percent change of the emission induced long term variation (E) by using OZIPR and the long term variation (L), and relative contribution of regional background ozone concentration change (B) are shown in Fig. 4 and Table 5. The L has increased about 14% from 2002 to 2006. On the contrary, the E showed a decreasing trend till 2005 (–20.9%) and then increased in 2006 compared to previous years but
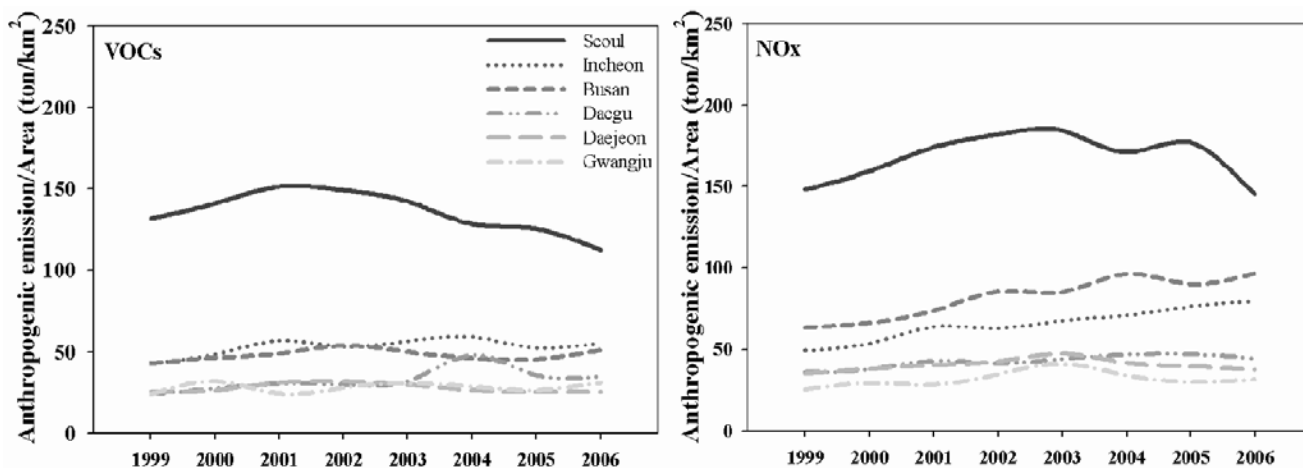

Fig. 3. Annual emission of ozone precursors in 6 cities: (a) Volatile organic compounds, and (b) nitrogen oxides.
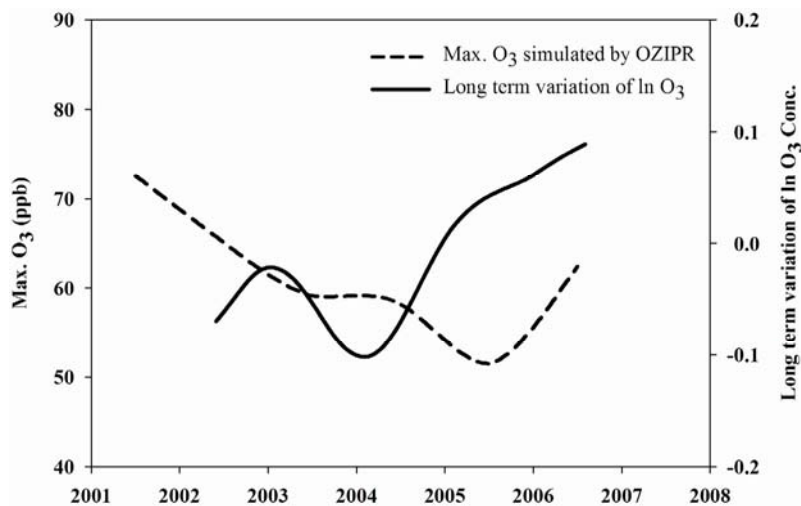

Fig. 4. Comparison of long term variation of ozone and simulated daily maximum ozone by OZIPR.

Table 5. The percent change of long term variation of ozone (L), the domestic emission-component long term variation of ozone (E), and relative contribution of regional background ozone concentration change (B).

Class \ Year	2002	2003	2004	2005	2006
L	100.0	103.9	97.0	108.2	113.6
E	100.0	90.8	89.4	79.1	95.9
B	-	13.1	7.6	29.1	17.7

The percent change of long term variation was calculated with the base year data (12th August, 2002), which is the first long term variation data extracted by KZ-filter method, and the data at the same day in every year. The percent change of the domestic emission-component long term variation of ozone was calculated with the simulated daily maximum ozone concentration induced by annual emission change in every year. The relative contribution of regional background ozone concentration change was inferred to be the difference between L and E.

still lower than 2002 (–4.1%). Generally, the B has increased more than 10% during the period. The difference between L and E was inferred to be the relative contribution of regional background ozone concentration change (B). The difference showed increasing trend with gradient 5.1 by the trend analysis. Thus it could be deduced that the contribution of regional background ozone concentration to long term variation of ozone has been increased. The relative importance of E and B was assumed to be represented by gradient calculated by the trend analysis of each component. The gradients of L, E, and B are 3.1, –2.0, and 5.1, respectively. Thus, the regional background effect is about 2.5 times more important than precursor emission in Seoul for the long term variation of the surface ozone concentration.

The background ozone concentrations of northern hemisphere are influenced from East Asian emissions (Fiore *et al.*, 2001). According to the annual report published by British Petroleum (BP), the fossil fuel consumption in Asia has increased continuously from 1999 up to 2009 (BP, 2010). If this increasing trend is continued in the future, the regional background effect might be more significant. Furthermore, the separation of long term variation of ozone into the components by domestic precursor emission change and regional background ozone concentration is to be more important for providing precise information to policy makers deciding emission control policy in Northeast Asia.

Short Term Change Due to Episodic Long Range Transport

Short term variation of ozone was also separated to the combined several possible factors affecting the ozone short term variation. Because the life time of ozone in the free troposphere is order of 1–2 months, ozone is able to transport over long distances (Gryning and Schiermeier, 2001). A clear example presenting long range transport of ozone over intercontinental from North American continent to Europe was presented by Stohl and Trickl (1999). Several other studies also showed the patterns of long range transport of ozone including the transportation in the Asian continent (Cristofanelli and Bonasoni, 2009; Ding *et al.*, 2009; Ghude *et al.*, 2010; Huang *et al.*, 2010; Junker *et al.*, 2009; Oh *et al.*, 2010).

In this study, it was assumed that the hourly and daily variation, and other short term variation like a episodic local emission change effect and episodic long range transport on atmospheric surface ozone concentration were

all dissolved in short term variation under 50 days, $O_{3ST}(t)$. Then, hourly variation couldn't be detected in this study because the resolution of data was on a daily basis. To distinguish the episodic long range transport, contour plots for short term variation of the ozone concentration with wind direction as x axis and wind speed as y axis were produced as shown in Fig. 5 for each city. Short term variation was mainly divided into two groups; high wind speed and low wind speed groups in all 6 cities. In case of Incheon, high short term variation along with full range of wind speed was observed in south-easterly wind and north-easterly wind direction. Because Seoul is located in the range from south-easterly to north-easterly direction of Incheon, it is inferred that the local emission change effect and episodic transport of ozone from Seoul, not long range transport form Asian continent, have effect on the short term variation of ozone concentration in Incheon.

Because the increase of air pollutant concentration induced by episodic long range transport is generally occurred under high wind speed and westerly wind condition in Korea (Han *et al.*, 2006), the high wind speed group with westerly wind direction was inferred as the signal of episodic long range transport. However, the high wind speed group with westerly wind direction was not clearly observed in cities, except for Daejeon and Gwangju. It is inferred that the local emission effect is relatively smaller in these two cities than other cities as shown in Fig. 3, so the signal of long range transport could be extracted.

Although it was not a quantitative result, the episodic long range transport and local emission effect of ozone could be separated from the original short term ozone trend qualitatively by using the low-pass KZ-filter method and simple graphical tool. Statistical method such as cluster analysis could be also applied to distinguish the various signals of short term variation. The quantitative separation of short term variation signal and quantification of episodic long range transport of ozone are intended to be studied in the future study.

SUMMARY AND CONCLUSIONS

The meteorologically adjusted long term variations of the daily 1-h maximum ozone concentration over 6 major cities in Korea were determined by using the low-pass KZ-filter and multiple linear regression methods.
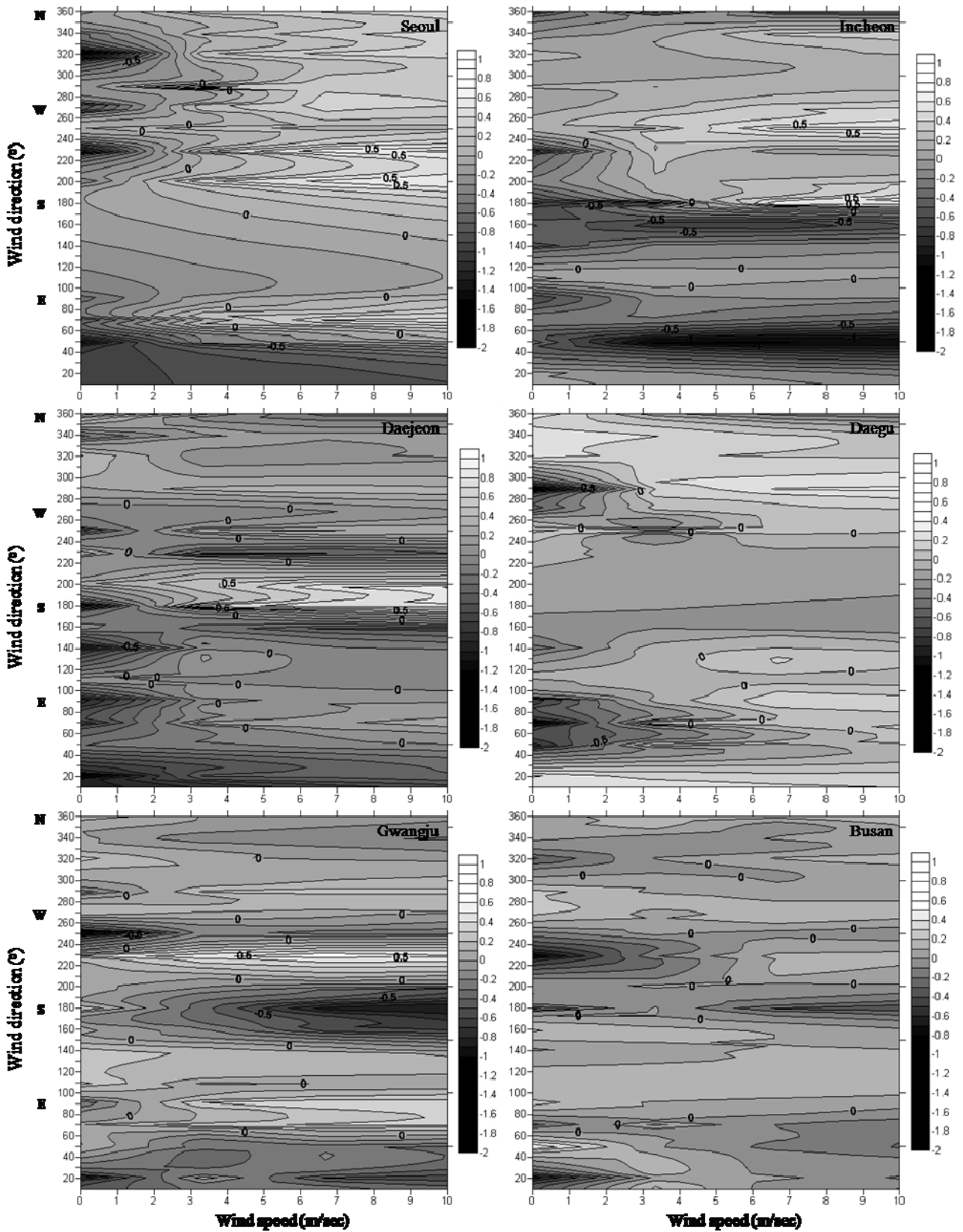
Fig. 5. Contour plot among short term variation of ozone, wind direction, and wind speed.

Generally, long term variation of ozone concentrations over Korea has increased by 3.3%/y (Seoul,), 2.2%/y (Gwangju), 1.1%/y (Daegu) during the study period except for Busan (–3.3%/y), Daejeon (–3.3%/y), and Incheon (–1.1%/y). The

trend of long term ozone variation in Korea is comparable with the results of other Asian region such as Beijing (2.0%/y) and Tokyo (about 1.0%/y). The seasonal variation was similar in all cities; high in spring and low in winter due to photochemical production in spring with the highest variation in Seoul. The meteorological factors could explain a major portion of the total variation of ozone concentration from 66.4% (Busan) to 89.7% (Seoul) based on the step-wised multiple linear regressions. Solar radiation was the strongest impacting meteorological factor correlated with ozone, showing high r^2 value from 0.647 (Busan) to 0.800 (Daegu). Ambient temperature was the second major factor affecting on the ozone concentration in Seoul, Incheon, and Daegu, but, relative humidity and wind speed were the second major factors in other three cities.

Long term variation in Korea was thought to be divided to two sub-components; the regional background ozone variation and the domestic precursor emission induced variation in this study. By using OZIPR model, the variation induced by the change of precursor emissions was separated from long term variation of ozone over Seoul, thus the effect of regional background ozone concentration was deduced. The percent change of the long term variation (L) of ozone has increased about 14% from 2002 to 2006. On the contrary, the percent change of the domestic precursor emission induced variation (E) showed a decreasing trend till 2005 (–21%) and then increased compared to previous years (–4.1%). The difference between L and E (7.6–29.1%) was inferred to the relative contribution of regional background ozone concentration change (B) to the long term variation of ozone. As a result of the trend analysis of L, E, and B, the L is thought to be accounted for about 75% by B and 25% by E for long term change. In other words, the regional background effect is about 2.5 times more important than precursor emission in Seoul.

Short term variation was qualitatively divided to the sub-components, induced by episodic long range transport (high wind speed group with westerly wind direction) and local emission, using contour plots for short term variation of ozone.

The results of this study are important because these can fill the gaps in the data in Northeast Asia by providing the recent 8 years ozone data. Also, by extracting the regional background effect, which is 2.5 times higher than emission effect in Seoul, from long term variation of ozone, the precise information can be provided to the policy makers by considering separated new impact factors and their effectiveness on emission control measures.

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