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The Long Term Trends of Tropospheric Ozone in Major Regions in Korea

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ABSTRACT

This study was conducted for analyzing the contribution factors on ozone concentrations and its long term trends in each major city and province in Korea through several statistical methods such as simple linear regression, generalized linear model, KZ-filer, correlation matrix, Kringing method, and cluster analysis. The overall ozone levels in South Korea have been consistently increasing over the past 10 years. The ozone concentrations in Seoul, the biggest city in Korea, are the lowest in all areas with the highest increasing ratio for 95^{th}% ozone. It is thought that the active photochemical reaction could affect the higher ozone concentration increase. On the other hand, the ozone concentrations in Jeju are the highest in Korea with the highest increasing ratio for 5^{th}%, 33^{th}%, and 50^{th}% ozone. It is also thought that the weak NO_x titration could be the reason of higher ozone concentrations in Jeju. In case of Jeju, transport related factors is the major factor affecting the ozone trend. Thus, it is assumed that the variation of ozone trend of Asian region affecting the ozone trend in Jeju, where domestic ozone photochemical reaction is less active than urban area. It is thought that the photochemical reaction plays the role of increasing of ozone concentrations in the urban area, even though the LRT affected on the increase of ozone concentrations in non-urban area.

Key words: Generalized linear model, Ozone trends, NO_x titration, Long range transport, Representative monitoring station

1. INTRODUCTION

Ozone is a major pollutant that has a significant impact not only on human activities and health but also on the ecosystem changes (Mauzerall *et al.*, 2005; Chameides *et al.*, 1999; Berntsen *et al.*, 1996). Recently, ozone levels in South Korea have been consistently rising, and frequency of high ozone concentrations have also continued to increase (KMOE, 2012). In particular, although public policies regulating emissions of ozone precursors such as volatile organic compounds (VOCs) and nitrogen oxides have been proposed to reduce ozone concentration, analysis of unintended factors contributing to ozone concentration such as meteorological factor or long range transport (LRT) have not yet been completed. Ozone is a secondary air pollutant formed by photochemical reaction. This is the primary reason for concentration level changes caused mainly by meteorological factors. LRT of ozone from windy regions such as China is recognized as one of the factors causing increased pollution in the Korean Peninsula (Kang *et al.*, 2013; Park *et al.*, 2012; Ghim, 2011). According to previous research, the region most affected by Asian air pollution is centered near 30 degrees and 40 degrees latitude, where South Korea is located (Tanimoto *et al.*, 2005). As such, the background concentrations of ozone in Korea are on the constant rise nationwide. Also, ozone concentration in Seoul has increased at higher rates than those in the United States, and it was found that increase in background ozone concentrations in Northeast Asia needs to be considered for estimating the ozone levels in South Korea (Tanimoto *et al.*, 2005; Vingarzan, 2004; Korsog and Wolff, 1991). According to the previous study, rising Asian anthropogenic emissions can increase the baseline ozone concentration even in U.S. (Jacob *et al.*, 1999).

Therefore, this study evaluates the impact of LRT of ozone and impact of meteorological factors in order to examine trends in ozone levels in South Korea, and also assess the factors contributing to changes in ozone level in each region. First, 16 representative long-term ozone monitoring stations were selected in 16 cities and province by applying statistical methods such as Kolmogorov-Zurbenko filter (KZ-filter), correlation matrix, cluster analysis, and Kriging method. Second, a basic statistical analysis such as calculating average and percentile concentration of ozone was done as well as simple linear regression for understanding the general characteristics of ozone long term trends in each cities and province. Finally, main factors causing fluctuations in ozone concentration in each city and province have been determined by applying the generalized linear model (GLM) to the selected representative monitoring stations. Also, the impact of precursor emissions and LRT on ozone concentration has been analyzed by examining the variation in ozone concentration.

It is imperative to analyze the contribution of unintended factors in Northeast Asia where LRT of air pollutants occurs frequently for understanding of the current status of air pollution. Also, policy-makers can use it as a reference when they establish policy goals to reduce ozone concentration. Therefore, the results of this study are meaningful not only for understanding the current pollution problems but also for establishing the future policy directions.

2. MATERIALS AND METHODS

2.1 Data Acquisition

The ozone data analyzed in this study have been measured at air quality monitoring stations nationwide by UV photometric method since 1989. The data used in the study were collected from monitoring stations in 16 cities and province for recent 10 years: Seoul (16 stations), Incheon (6 stations), Busan (10 stations), Daegu (4 stations), Daejeon (3 stations), Gwangju (4 stations), Gyeonggi (28 stations), Ulsan (7 stations), Gangwon (3 stations), Gyeongbuk (10 stations), Gyeongnam (7 stations), Jeonbuk (4 stations), Jeonnam (6 stations), Chungbuk (3 stations), Chungnam (3 stations), Jeju (3 stations). The locations of each atmospheric monitoring station in 16 the cities and provinces are shown in Fig. 1. More detailed information including longitude and latitude for each monitoring station are shown in Table S1. The hourly ozone concentrations at each monitoring station collected and managed by the National Air Quality Data Center (NAQDC), National

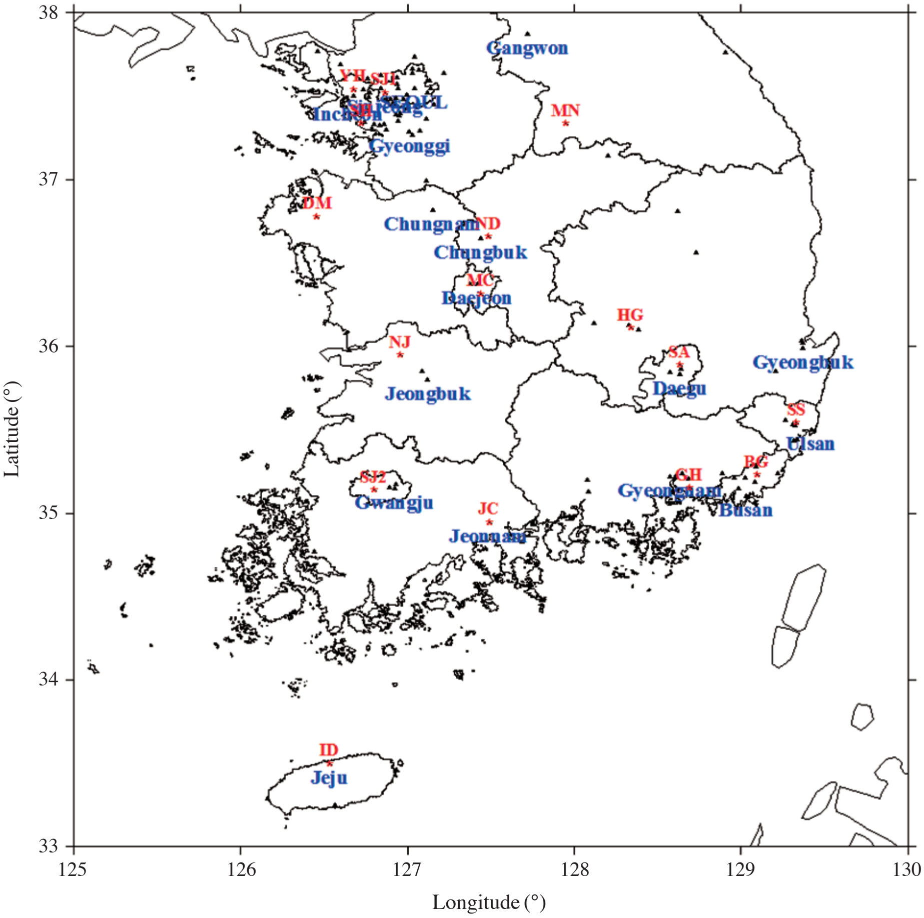

Fig. 1. The locations of each atmospheric monitoring station and a representative monitoring station in 16 cities and provinces.

Table 1. The list of parameters considered as input of adjustment model.

Parameters	Comments
8-hr maximum O_3 concentration (ppm)	
Daily maximum temperature (T_{max}, °C)	
Daytime average humidity (RH, %)	10 am-4 pm average
Morning average wind speed (WS_m, m s^{-1})	7-10 am
Afternoon average wind speed (WS_a, m s^{-1})	1-4 pm
Morning surface temperature difference (T_{ds}, °C)	Temperature at 925 mb - temperature at surface at 12 pm
Deviation in 1200 UTC temperature (T_{d850}, °C)	850 mb surface from 10 - year monthly average
Transport direction (T_{Dis}, °)	Degrees clockwise from North
Transport distance (T_{Dir}, km)	
Duration of precipitation (H_{rain}, hr)	

Institute of Environmental Research (NIER), were used to calculate daily 1 hr maximum concentrations by city from 2002 to 2011. Every 1 hr ozone concentration at each station was averaged to determine 1 hr ozone concentrations and daily 1 hr maximum concentrations for each city. The time scale used in the ozone trend analysis varied from 5 minutes to a day, but daily 1 hr maximum concentration was most widely used in the previous studies (Thompson *et al*., 2001). Also, according to the human health and air pollution related studies, acute exposal to ozone cause several health effects such as arterial vasoconstriction, heart rate decreasing, pulmonary function, mortality (Brook *et al*., 2002; Gold *et al*., 2000; Folinsbee *et al*., 1988; Touloumi *et al*., 1997). Therefore, daily 1 hr maximum concentration was used in this study for further analyses except for general statistical analysis. Meteorological data at the Regional Meteorological Observatory for each city and province were obtained from the Korea Meteorological Administration at the same resolution for the same time period. Meteorological factors of interest were daily maximum ambient temperatures (T_{max}), mid-day average relative humidity (RH), morning and afternoon average wind speeds (WS_m and WS_a), morning surface temperature differences (T_{ds}), deviation in temperature from a 10-year monthly mean at 850 mb (T_{d850}). These factors were the most important meteorological variables selected by statistical analysis between ozone concentration and various meteorological sub-parameters. LRT factors of interest were 24 hr HYSPLIT transport directions and distances (T_{Dir} and T_{Dis}) as shown in Table 1. Details on the related methods were described in Camalier *et al.* (2007). Some of these variables were directly observed, while others were calculated based on observed data or other parameters. In addition, transport directions and distances were determined based on the hybrid single-particle lagrangian integrated trajectory (HYSPLIT) model simulations (Draxlerand Hess, 1997). The HYSPLIT model was run to calculate 24 hr backward trajectories from each surface station. The trajectories were started at noon LST at a height of 300 m (i.e., within the mixed layer). The statistical summary of the meteorological and LRT parameters are given in Table 2.

2. 2 Statistical Methods

To investigate the characteristics of ozone trends during a whole year including each season (spring, summer, fall and winter), the seasonal 5th%, 33th%, 50th%, 67th%, and 95th% ozone concentration were considered. The ozone rate of change (ppb per year) over 2002-2011 was calculated separately for each ozone percentile through the simple linear regression. The trends were calculated with a reference year of 2002.

In previous studies, various statistical methods have been applied to analyze the long term variation of tropospheric ozone concentration such as time series decomposition, KZ-filter, GAM, DLM, and GLM (Shin *et al*., 2012; Development Core Team, 2009; Lee *et al*., 2008; U.S. EPA, 2002; R Schmidt, 2001; Cressie, 1993; Hastie *et al*., 1990). Because the long term trends of ozone is influenced by meteorological conditions, it is important to deduce the long term trends excluding changing meteorological factors. Many previous studies attempted to infer the actual fluctuations of ozone levels by substituting meteorological factors (Camalier *et al*., 2007; Zheng *et al*., 2007; Thompson *et al*., 2001; Davis *et al*., 1998; Bloomfield *et al*., 1996). GLM is a large class of statistical model for linear combination of expected variables including dependent variables and common errors. GLM explains the relationships between variables selected from urban ozone and

Table 2. Statistical summary of the meteorological and LRT parameters.

	T_{max} (°C)	RH (%)	WS_m (m/s)	WS_a (m/s)	T_{ds} (°C)	T_{d850} (°C)	T_{Dir} (km)	T_{Dis} (°)	Hrain (hour)
Seoul	26.4 ± 3.7	58.8 ± 18.1	2.1 ± 1.0	2.9 ± 1.1	−5.6 ± 2.2	0.0 ± 2.7	400.9 ± 207.3	166.7 ± 108.9	4.3 ± 6.4
Busan	25.3 ± 3.7	68.1 ± 14.5	2.1 ± 1.2	3.1 ± 1.3	−5.9 ± 3.2	0.0 ± 3.0	465.4 ± 236.7	168.9 ± 109.9	3.6 ± 5.7
Daegu	27.9 ± 4.2	54.6 ± 17.8	1.8 ± 1.1	3.2 ± 1.2	−7.6 ± 2.8	0.0 ± 3.0	428.9 ± 221.3	168.2 ± 108.5	3.9 ± 5.9
Incheon	25.4 ± 3.6	65.4 ± 16.8	2.3 ± 1.5	3.5 ± 1.5	−4.8 ± 2.5	0.0 ± 2.7	384.5 ± 207.0	169.9 ± 110.7	3.8 ± 6.0
Gwangju	27.5 ± 3.7	61.9 ± 17.0	3.3 ± 2.3	3.9 ± 2.1	−6.7 ± 2.2	0.0 ± 2.5	448.8 ± 245.4	175.9 ± 112.4	4.1 ± 5.9
Daejeon	26.8 ± 3.6	59.3 ± 17.4	1.8 ± 1.0	2.8 ± 1.1	−6.0 ± 2.2	0.0 ± 2.7	403.0 ± 218.4	169.2 ± 110.1	4.3 ± 6.1
Ulsan	26.4 ± 4.3	62.6 ± 16.4	2.1 ± 1.5	3.1 ± 1.6	−6.8 ± 2.8	0.0 ± 3.0	430.2 ± 223.8	168.0 ± 108.5	3.9 ± 6.0
Gyeonggi	26.6 ± 3.8	61.8 ± 17.5	1.6 ± 0.9	2.5 ± 1.0	−5.8 ± 2.2	0.0 ± 2.7	406.4 ± 209.9	167.5 ± 109.8	4.1 ± 6.2
Gangwon	25.1 ± 4.7	61.7 ± 19.2	2.0 ± 1.2	2.6 ± 1.1	−6.0 ± 2.9	0.0 ± 3.3	413.2 ± 208.0	160.7 ± 102.7	4.7 ± 6.7
Chungbuk	27.2 ± 3.6	57.7 ± 17.0	2.1 ± 1.0	2.9 ± 1.1	−6.1 ± 2.2	0.0 ± 2.7	396.2 ± 213.1	167.1 ± 109.4	3.9 ± 5.9
Chungnam	25.8 ± 3.7	66.0 ± 16.5	1.9 ± 1.3	2.8 ± 1.3	−5.4 ± 2.2	0.0 ± 2.7	392.5 ± 211.1	174.0 ± 112.9	3.7 ± 5.7
Jeonbuk	27.9 ± 3.7	57.2 ± 17.1	1.7 ± 1.1	3.1 ± 1.3	−7.2 ± 2.3	0.0 ± 2.5	423.0 ± 221.9	171.3 ± 111.0	3.7 ± 5.5
Jeonnam	24.8 ± 3.5	69.0 ± 16.0	1.9 ± 1.6	3.0 ± 1.5	−4.7 ± 2.2	0.0 ± 2.5	444.4 ± 238.8	175.2 ± 111.9	3.4 ± 5.7
Gyungbuk	26.0 ± 4.7	66.5 ± 16.9	1.6 ± 1.0	2.8 ± 1.1	−6.1 ± 2.8	0.0 ± 3.0	446.0 ± 229.9	165.5 ± 106.8	4.0 ± 6.1
Gyungnam	26.4 ± 3.8	64.2 ± 15.4	3.1 ± 2.0	3.9 ± 1.9	−6.9 ± 3.1	0.0 ± 3.0	445.1 ± 238.0	170.4 ± 110.1	3.9 ± 6.2
Jeju	26.0 ± 4.1	67.0 ± 13.2	1.3 ± 1.0	2.4 ± 1.2	−5.0 ± 2.7	0.0 ± 2.4	467.1 ± 249.0	187.3 ± 112.6	3.7 ± 5.7

broad-ranged meteorological variables (Camalier *et al.*, 2007). Because meteorological variables that affect ozone concentration as well as wind directions and distances calculated from the HYSPLIT model are used in GLM modeling, GLM can possibly diminish the impact of meteorological factors or LRT if these values are adjusted. In this study, these characteristics of GLM were used to investigate factors influencing the ozone trends.

GLM models used to evaluate meteorological adjusted ozone trends are generally expressed as follows:

$$g(\mu_i)=\beta_0+f(x_i,1)+\ldots fj(x_i,j)+..fp(x_i,p)_k+W_d+Y_k$$

Where, "i" represents the highest daily ozone concentration in an 8-hour period, "j" each meteorological factor, and "k" the year of the measurement. "β_0" represents the average value of total ozone, "f" the smoothing function related to "j", "W_d" days in the week, and "Y_k" the annual average concentration of ozone during the year "k" that takes into account meteorological conditions. "g" is the connecting function that represents the correlations between all variables on the right-hand side of the equation.

The meteorological factor is known to play an important role in long-term fluctuation of pollution levels (Kong *et al.*, 2006; Wise and Comrie, 2005; Dueñas *et al.*, 2002; Abdul-Wahab *et al.*, 1996; Cox and Chu, 1993). In particular, concentrations of ozone formed through photochemical reactions fluctuate significantly depending on meteorological factors such as temperature, wind direction, and stability. If only ozone level is taken into account, periodic fluctuations due to meteorological factors, LRT, and precursor emissions will comprehensively impact the long term trends. Since GLM can be used to selectively remove meteorological factors and movement in air parcels in the long term trends, and only consider the impact of LRT or precursor emissions, GLM was used to analyze the long term trends in ozone concentration.

2. 3 Selection of Representative Monitoring Stations in Each City and Province

As shown in Fig. 1, there are many sampling sites in each city and province. To characterize the long term trends of ozone by area, it is difficult to deal with all data in individual monitoring station. Thus, representative monitoring station was selected and their data was used for further trend analysis of each city and province. Among atmospheric monitoring stations in each cities and province, stations that have sufficient data with data continuity for the period from 2002 to 2011 were selected, and ozone concentration data collected from the stations in each city and province were analyzed using KZ-filter, correlation matrix, cluster analysis, and Kriging method for selecting representative monitoring station.

Firstly, KZ-filter was used to calculate annual changes in each monitoring station after eliminating seasonal fluctuations and random variables. Annual changes, which were extracted by KZ-filter, in each monitoring station were compared with those of other monitoring stations within same city and province to check similarities with other stations by correlation analysis. And the result was displayed in a correlation matrix. Additionally, to examine the spatial distribution of ozone annual changes, Kriging method was applied. Also, cluster analysis is used for identifying the homogenous groups of monitoring stations based on their similarity of annual changes. Along with the results from cluster analysis, correlation matrix, and Kriging method, representative monitoring stations, where correlation coefficient was high in the largest cluster group with spatially homogeneity, in each city and province were selected.

3. RESULTS

3. 1 Selection of Representative Monitoring Stations

Among the 16 cities and province, the process of selecting representative monitoring stations in Seoul is detailed in Shin *et al.* (2015). Summary of the methodology used for selecting the station in Seoul is as follows. A total of 16 monitoring stations that had accumulated data over 10 years were selected out of the 25 monitoring stations in Seoul. In order to select representative monitoring stations, KZ-filter was applied in calculating the annual changes in each monitoring station from the time-series data composed of complicated cycless after excluding seasonal changes and random variables from short-term fluctuations. After calculation of the actual long term fluctuation patterns using the KZ-filter method, the correlation among each monitoring station was analyzed and compared to each other. The correlation between monitoring stations was somewhat high, ranging from 0.52-0.87. The highest correlation was observed at Sinjeong monitoring station (0.87) followed by Sadang (0.86) and Beon (0.82). Together with the correlation matrix, cluster analysis can be used to select representative monitoring stations by grouping several monitoring stations in each region. Based on the cluster analysis, 16 monitoring stations in Seoul were grouped into 4 clusters. Kriging method linearly combines actual values for interpolation, and when predicting values, the method reflects not only the deviation from the actual values but also the correlation coefficient with each neighboring value. Therefore, the method was used to select representative monitoring stations based on the interpretation of the spatial distribution of ozone concentrations. The results are similar to those of correlation matrix calculated by applying KZ-filter as well as the cluster analysis. Correlations among Sinjeong, Bangi, and Beon showed the highest spatial distribution when the Kriging method was applied.

The various methods applied to select representative monitoring stations in Seoul show similar results. The result of correlation matrix shows high correlations of Sinjeong, Sadang, and Beon. Based on the cluster analysis, 16 monitoring stations in Seoul were grouped into 4 clusters. Sinjeong, Sadang, and Beon formed a cluster. Similar to the above-mentioned two methods, the Kriging method again shows high correlation of Sinjeong, Bangi, and Beon.

Table 3. The representative stations in each city and province.

City and Province	Sites	Averaged correlation coefficient
Seoul	Sinjeong (SJ1)	0.87
Busan	Bugok (BG)	0.53
Daegu	Sinam (SA)	0.78
Incheon	Yeonhui (YH)	0.36
Gwangju	Songjeong (SJ2)	0.93
Daejeon	Munchang (MC)	0.86
Ulsan	Samsan (SS)	0.58
Gyeonggi	Sihwa (SH)	0.64
Gangwon	Myeongnyun (MN)	0.73
Chungbuk	Naedeok (NG)	0.38
Chungnam	Dongmun (DM)	0.53
Jeonbuk	Namjung (NJ)	0.72
Jeonnam	Jangcheon (JC)	0.39
Gyeongbuk	Hyeonggok (HG)	0.28
Gyeongnam	Gyeonghwa (GH)	0.37
Jeju	Ido (ID)	0.79

The study found that the station at Sinjeong represented the long term trends of ozone better than 15 other monitoring stations. For other cities and province, the same procedure was applied and some of the results are shown in Fig. S1 and Fig. S2. Table 3 shows representative monitoring stations selected along with the correlation with other stations within each city and province after applying the above-mentioned methods to the monitoring stations in the 16 cities and province. Fig. 1 also shows locations of representative atmospheric monitoring stations in 16 cities and province.

3. 2 The Characteristics of Ozone Concentrations

Annual average ozone concentrations calculated by hourly ozone concentrations including night time data in the 16 cities and provinces are given in Table 4. As shown in Table 4, the annual average concentration of ozone in 16 cities ranged between 0.014 ppm and 0.043 ppm for the period with concentration increasing from 0.022 ppm in 2002 to 0.025 ppm in 2011. The 10 year-average ozone concentration was the lowest at 0.018 ppm and the highest at 0.036 ppm in Seoul and Jeju, respectively. Jeju is the background area surrounded by sea with lowest population density and vehicle registration, whereas Seoul is the biggest city with the largest vehicle registrations in Korea (KOSIS, 2014). Thus, it is assumed that the ozone concentrations in Seoul are the lowest because of NO_x titration. The spatial distributions of ozone percentile concentration for

Table 4. Annual average concentrations of atmospheric ozone in 16 cities and provinces in Korea between 2002 and 2011. (unit: ppm)

	2002	2003	2004	2005	2006	2007	2008	2009	2010	2011	Avg. by region
Seoul (SE)	0.014	0.014	0.014	0.018	0.018	0.018	0.019	0.022	0.020	0.020	0.018
Busan (BS)	0.024	0.023	0.024	0.022	0.022	0.023	0.025	0.025	0.025	0.026	0.024
Daegu (DG)	0.019	0.021	0.023	0.024	0.020	0.022	0.022	0.023	0.022	0.025	0.022
Incheon (IC)	0.021	0.022	0.022	0.023	0.021	0.023	0.024	0.024	0.021	0.022	0.022
Gwangju (GJ)	0.016	0.018	0.022	0.023	0.022	0.022	0.021	0.025	0.023	0.025	0.022
Daejeon (DJ)	0.019	0.019	0.020	0.022	0.020	0.018	0.024	0.023	0.023	0.022	0.021
Ulsan (US)	0.019	0.020	0.022	0.022	0.021	0.021	0.023	0.025	0.023	0.026	0.022
Gyeonggi (GG)	0.017	0.018	0.019	0.019	0.019	0.019	0.020	0.022	0.021	0.021	0.020
Gangwon (GW)	0.023	0.022	0.023	0.023	0.024	0.024	0.026	0.027	0.024	0.025	0.024
Chungbuk (CB)	0.021	0.022	0.021	0.021	0.022	0.023	0.021	0.024	0.022	0.023	0.022
Chungnam (CN)	0.026	0.024	0.024	0.023	0.021	0.020	0.023	0.026	0.027	0.024	0.024
Jeonbuk (JB)	0.019	0.017	0.017	0.018	0.020	0.018	0.023	0.024	0.024	0.022	0.020
Jeonnam (JN)	0.026	0.031	0.032	0.027	0.025	0.030	0.030	0.030	0.026	0.028	0.029
Gyeongbuk (GB)	0.023	0.024	0.024	0.025	0.024	0.024	0.025	0.026	0.024	0.025	0.024
Gyeongnam (GN)	0.025	0.026	0.026	0.024	0.027	0.027	0.028	0.028	0.025	0.026	0.026
Jeju (JJ)	0.034	0.031	0.033	0.033	0.035	0.035	0.038	0.043	0.041	0.037	0.036
Avg. by year	0.022	0.022	0.023	0.023	0.023	0.023	0.025	0.026	0.024	0.025	

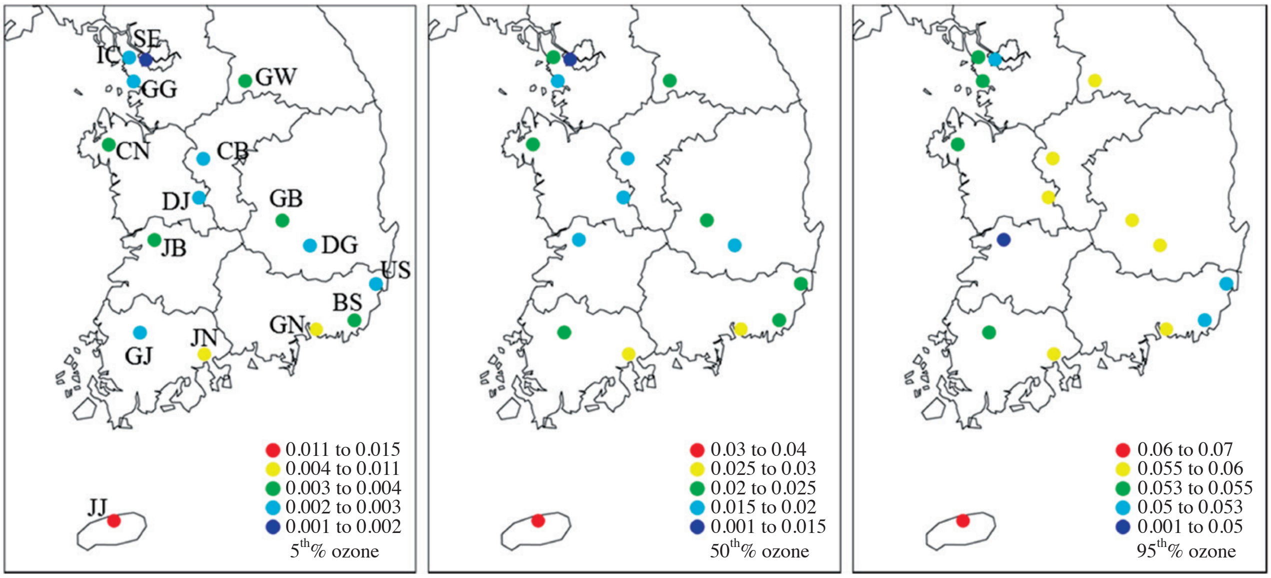

Fig. 2. The spatial distribution of 5th%, 50th%, and 95th% ozone concentration for 2002-2011.

10 year are shown in Fig. 2. For 5th%, 50th%, 95th% ozone concentration, Jeju shows the highest concentration among all other area. For 5th% and 50th% ozone, south coastal areas show relatively high ozone concentration. However, for 95th% ozone, inland areas show relatively high ozone concentration as well as south coastal area. Seoul metropolitan areas show relatively lower ozone concentration than other area. NO_x emissions for Seoul and Jeju for 2001 are shown in Table 5. NO_x emissions in Seoul are 4.3-10.0 times higher than those in Jeju. As mentioned above, it is assumed that the NO_x titration cause the ozone concentration difference between two sites.

The national air quality standards for ozone are 0.06 ppm for 8 hours (99 percentile) and 0.10 ppm for 1 hour (999 per mill) in Korea. Considering the annual average ozone concentrations (0.007-0.026 ppm) in the UK in 2012 and the daily maximum ozone concentration (0.024 ppm) in Spain for 10 years (2001-2010), ozone concentrations are relatively high in Korea

Table 5. Annual NO_x emissions between 2003 and 2011 for Seoul and Jeju. (unit: ton/year)

	2003	2004	2005	2006	2007	2008	2009	2010	2011
Seoul (SE)	111,698	103,549	107,257	87,893	113,086	71,493	66,998	71,070	62,067
Jeju (JJ)	13,001	12,589	12,304	11,632	11,270	9,568	10,140	11,786	14,549

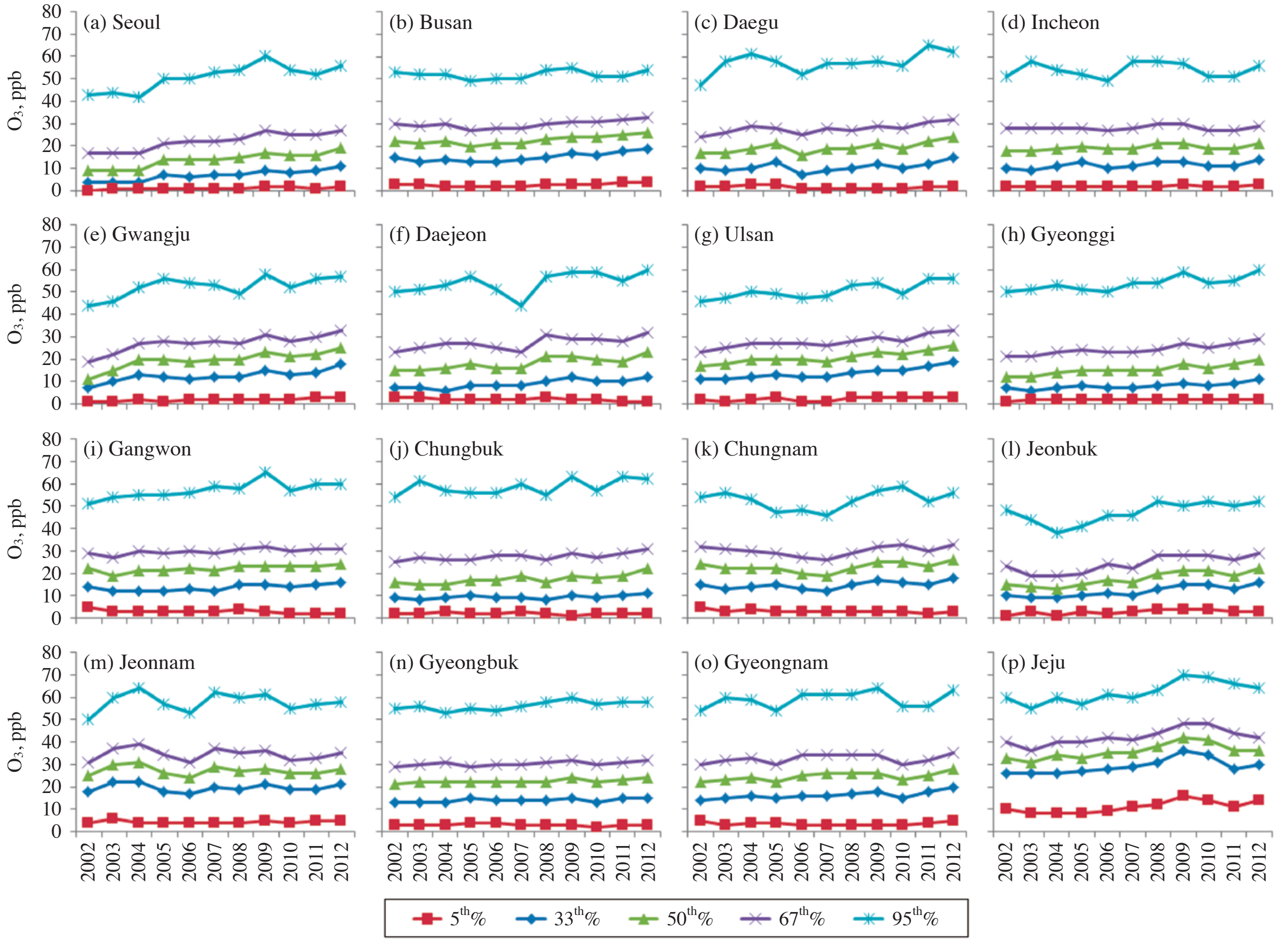

Fig. 3. Long term trends of percentile ozone of whole year in each city and province (dark pink: 5th%; blue: 33th%, green: 50th%, purple: 67th%, sky blue: 95th%).

(Santurtún *et al*., 2015; DEFRA). However, ozone concentrations in Korea are slightly lower than those in China. In previous studies, surface ozone concentration varied from 0.016 to 0.053 ppm (monthly median) and from 0.018 to 0.047 ppm in eastern China (1991-2006) and southern China (1994-2007), respectively (Wang *et al*., 2009; Xu *et al*., 2008). Long term variations of ozone for each city and province of the whole year and each season by each percentile are shown from Fig. 3 to Fig. 7. And, ozone rate of changes are listed up in Table 6. As shown in Fig. 3 and Table 6, ozone rate of changes for the whole year are −0.04 (IC) - 1.55 (SE) ppb yr^{-1} for 95th% ozone, −0.14 (JN) - 1.12 (SE) ppb yr^{-1} for 67th% ozone, −0.16 (JN) - 0.95 (GJ) ppb yr^{-1} for 50th% ozone, −0.07 (JN) - 0.83 (JJ) ppb yr^{-1} for 33th% ozone, −0.19 (GW) - 0.64 (JJ) ppb yr^{-1} for 5th% ozone. Considering the whole year ozone concentration changes, Seoul has the highest increase ratio for higher ozone concentration (95th%, 67th%). On the other hand, IC is the lowest increasing area for the highest ozone concentration (95th%) and JN is the lowest increasing area for mid-range ozone concentration (67th%, 50th%, and 33th%). JJ is the highest increasing area for lower ozone concentration (33th% and 5th%).

Table 6. Ozone rate of changes over 2002-2012 for each city and province by each ozone percentile. (unit: ppb/year)

		SE	BS	DG	IC	GJ	DJ	US	GG	GW	CB	CN	JB	JN	GB	GN	JJ	Range
Whole year	95%	1.55	0.03	0.82	−0.04	0.96	0.75	0.83	0.66	0.99	0.51	0.22	1.01	0.19	0.48	0.22	1.33	−0.04-1.55
	67%	1.12	0.28	0.47	0.01	0.96	0.56	0.72	0.62	0.33	0.31	0.03	0.95	−0.14	0.18	0.13	1.04	−0.14-1.12
	50%	0.94	0.41	0.40	0.18	0.95	0.64	0.65	0.62	0.32	0.41	0.16	0.84	−0.16	0.17	0.30	0.92	−0.16-0.95
	33%	0.60	0.42	0.17	0.21	0.58	0.51	0.61	0.25	0.28	0.09	0.21	0.65	−0.07	0.15	0.32	0.83	−0.07-0.83
	5%	0.13	0.10	−0.14	0.03	0.17	−0.13	0.17	0.05	−0.19	−0.05	−0.19	0.27	0.00	−0.07	−0.10	0.64	−0.19-0.64
Spring	95%	1.67	0.03	0.70	−0.13	1.18	0.75	0.45	0.58	0.94	0.45	0.14	0.75	0.65	0.61	0.01	1.87	−0.13-1.87
	67%	1.77	0.16	0.68	0.02	1.38	0.58	0.75	0.80	0.78	0.40	0.18	0.96	0.36	0.52	0.16	1.54	0.02-1.77
	50%	1.81	0.37	0.95	0.03	1.35	0.82	0.87	1.13	0.87	0.79	0.32	1.10	0.26	0.75	0.35	1.58	0.03-1.81
	33%	1.59	0.67	1.12	0.38	1.32	1.22	0.96	1.03	0.91	1.11	0.52	1.13	0.28	0.99	0.44	1.49	0.28-1.59
	5%	0.22	0.35	0.01	0.15	0.35	−0.03	0.15	0.08	0.35	0.22	0.03	0.55	0.50	0.18	0.22	1.29	−0.03-1.29
Summer	95%	1.79	−0.12	1.26	0.15	0.78	1.25	1.23	1.02	1.60	1.11	0.36	1.35	−0.24	0.58	0.01	0.68	−0.24-1.79
	67%	1.15	0.13	0.61	0.23	1.04	1.38	0.86	0.88	0.85	1.01	0.33	1.12	−0.18	0.25	0.24	0.73	−0.18-1.38
	50%	0.92	0.27	0.37	0.32	0.84	1.19	0.73	0.67	0.64	0.85	0.25	0.87	−0.18	0.17	0.25	0.56	−0.18-1.19
	33%	0.61	0.31	0.31	0.18	0.61	0.97	0.55	0.38	0.51	0.47	0.13	0.70	−0.11	0.09	0.39	0.49	−0.11-0.97
	5%	0.22	0.33	−0.16	0.07	0.43	−0.02	0.37	0.13	0.03	0.04	−0.14	0.33	−0.03	−0.06	0.13	0.58	−0.16-0.58
Fall	95%	1.07	0.17	1.25	0.12	0.89	0.41	1.26	0.70	0.47	−0.02	0.28	1.58	0.05	0.24	0.91	0.78	−0.02-1.58
	67%	0.79	0.39	0.53	0.05	0.82	0.40	0.89	0.64	0.22	−0.02	−0.07	0.82	−0.30	0.02	0.28	0.68	−0.30-0.89
	50%	0.59	0.52	0.48	0.24	0.84	0.45	0.80	0.44	0.35	−0.02	0.02	0.81	−0.27	0.01	0.28	0.67	−0.27-0.84
	33%	0.19	0.59	−0.21	0.15	0.42	0.05	0.50	0.03	0.16	−0.28	0.19	0.63	−0.11	−0.11	0.21	0.80	−0.28-0.80
	5%	0.08	0.07	−0.15	0.12	0.18	−0.07	0.15	0.13	−0.21	−0.11	−0.27	0.28	−0.04	−0.11	−0.08	0.81	−0.27-0.81
Winter	95%	0.90	−0.30	−0.59	−0.36	0.41	−0.82	−0.21	0.21	−0.29	−0.58	−0.45	0.23	−0.34	−0.26	−0.38	1.01	−0.82-1.01
	67%	0.82	0.06	−0.05	−0.19	0.90	−0.06	0.18	0.40	−0.20	−0.20	−0.17	0.78	−0.35	−0.05	−0.17	0.93	−0.35-0.93
	50%	0.52	0.43	0.02	−0.05	0.62	0.08	0.31	0.15	−0.16	−0.25	0.10	0.42	−0.32	0.07	0.13	0.88	−0.32-0.88
	33%	0.18	0.44	−0.20	−0.01	0.02	−0.07	0.38	−0.04	−0.14	−0.36	0.15	0.16	−0.25	0.09	0.18	1.01	−0.36-1.01
	5%	0.10	0.07	−0.09	0.00	0.16	−0.12	0.04	0.10	−0.19	−0.18	−0.22	0.12	−0.09	0.02	−0.09	0.70	−0.22-0.70

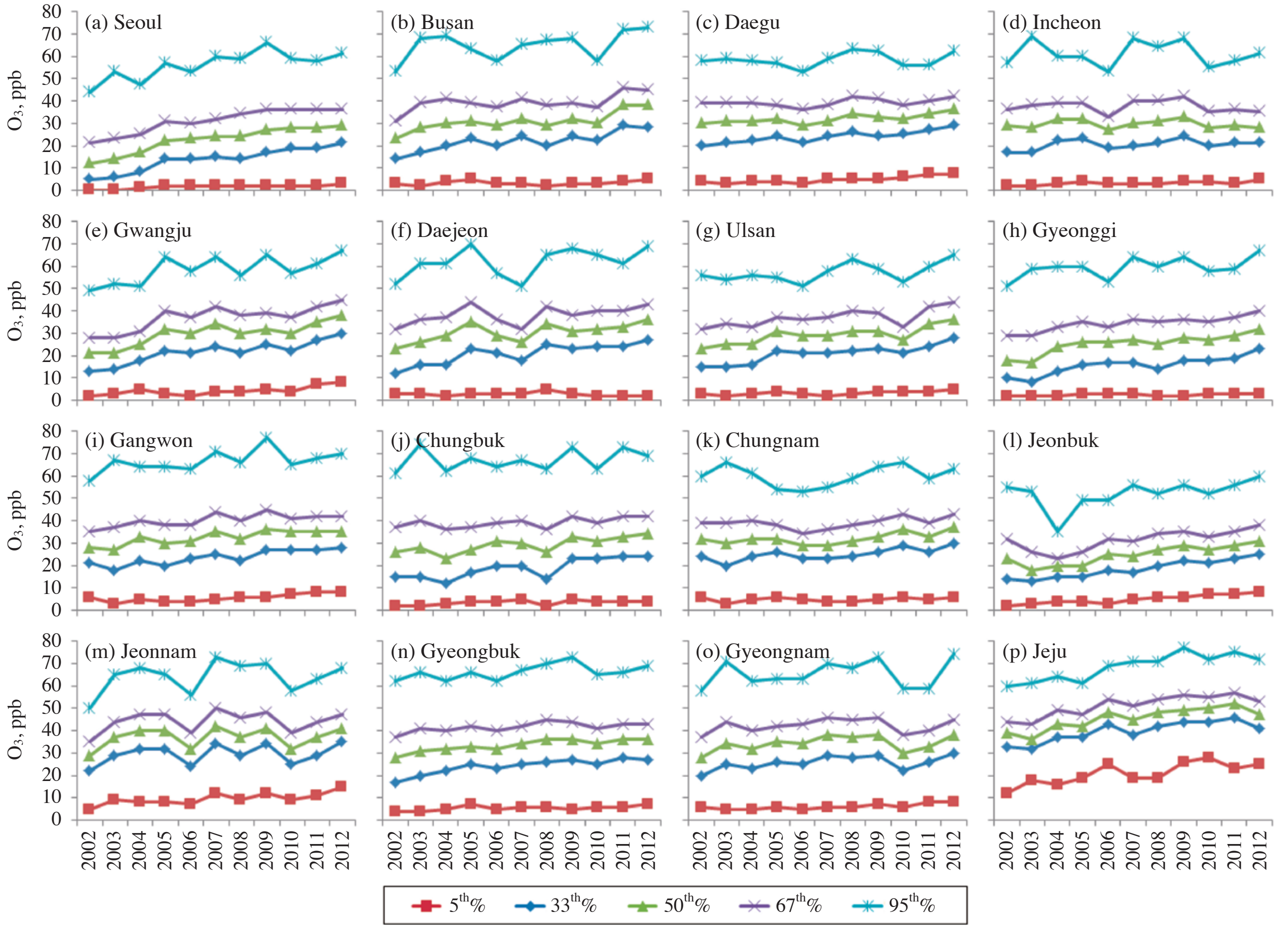

Fig. 4. Long term trends of percentile ozone of spring in each city and province (dark pink: 5th%; blue: 33th%, green: 50th%, purple: 67th%, sky blue: 95th%).

The changing ratio of ozone is from −0.13 ppb yr^{-1} (IC) to 1.87 ppb yr^{-1} (JJ) in spring, from −0.24 ppb yr^{-1} (JN) to 1.79 ppb yr^{-1} (SE) in summer, from −0.02 ppb yr^{-1} (CB) to 1.58 ppb yr^{-1} (JB) in fall, and from −0.82 ppb yr^{-1} (DJ) to 1.01 ppb yr^{-1} (JJ) in winter for 95th% ozone concentrations (Figs. 4-7, Table 6). For 67th% ozone concentrations, the changing ratio of ozone is from 0.02 ppb yr^{-1} (IC) to 1.77 ppb yr^{-1} (SE) in spring, from −0.18 ppb yr^{-1} (JN) to 1.38 ppb yr^{-1} (DJ) in summer, from −0.30 ppb yr^{-1} (JN) to 0.89 ppb yr^{-1} (US) in fall, and from −0.35 ppb yr^{-1} (JN) to 0.93 ppb yr^{-1} (JJ). For 50th% ozone concentrations, the changing ratio of ozone is from 0.03 ppb yr^{-1} (IC) to 1.81 ppb yr^{-1} (SE) in spring, from −0.18 ppb yr^{-1} (JN) to 1.19 ppb yr^{-1} (DJ) in summer, from −0.27 ppb yr^{-1} (JN) to 0.84 ppb yr^{-1} (GJ) in fall, and from −0.32 ppb yr^{-1} (JN) to 0.88 ppb yr^{-1} (JJ). For 33th% ozone concentrations, the changing ratio of ozone is from 0.28 ppb yr^{-1} (JN) to 1.59 ppb yr^{-1} (SE) in spring, from −0.11 ppb yr^{-1} (JN) to 0.97 ppb yr^{-1} (DJ) in summer, from −0.28 ppb yr^{-1} (CB) to 0.80 ppb yr^{-1} (JJ) in fall, and from −0.36 ppb yr^{-1} (CB) to 1.01 ppb yr^{-1} (JJ). For 5th% ozone concentrations, the changing ratio of ozone is from −0.03 ppb yr^{-1} (DJ) to 1.29 ppb yr^{-1} (JJ) in spring, from −0.16 ppb yr^{-1} (DG) to 0.58 ppb yr^{-1} (JJ) in summer, from −0.27 ppb yr^{-1} (CN) to 0.81 ppb yr^{-1} (JJ) in fall, and from −0.22 ppb yr^{-1} (CN) to 0.70 ppb yr^{-1} (JJ). According to Xu *et al.* (2008), long term trend of surface ozone in eastern China for 25 years (1991-2006, −0.79-−0.33 ppb yr^{-1}) has been decreasing at a moderate rate since the early 1990s. However, the ozone concentration increased by an average rate of 0.55 ppb yr^{-1} in the southern China (1994-2007) (Wang *et al.*, 2009). Even though long term trend of ozone are varied in China due to different status of economic development and related precursor emission within China, it was recognized that the precursor emission cause the changing of ozone long term trend. According to Cooper *et al.* (2012), spring season

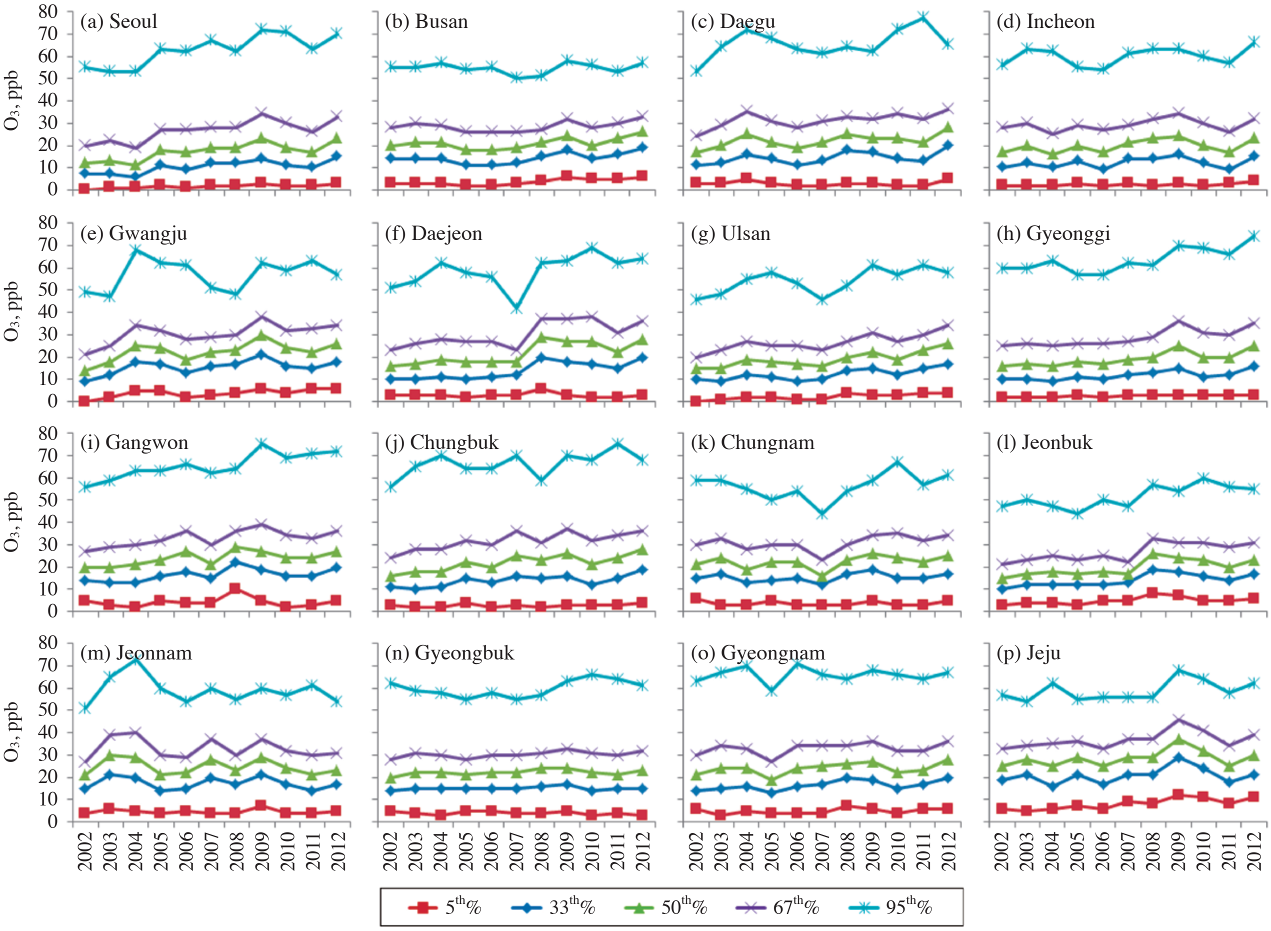

Fig. 5. Long term trends of percentile ozone of summer in each city and province (dark pink: 5th%; blue: 33th%, green: 50th%, purple: 67th%, sky blue: 95th%).

ozone trends above western North America for 1995-2011 (3-8 km above sea level) was −0.03-1.29 for 95th %, 0.15-0.71 for 67th%, 0.14-0.68 for 50th%, 0.12-0.66 for 33th%, −0.11-0.65 for 5th%. Considering this result, the ozone increasing ratio in the Korean Peninsula is higher than those in higher ozone areas in the US even though there are some differences by area. Also, considering the area with the highest increasing ratio (1.29-1.87 ppb yr^{-1}) for each percentile ozone concentration in spring, increasing ratio of ozone in the Korean Peninsula are higher than those reported for downwind of Asia (0.29-0.97 ppb yr^{-1}) reported by Cooper *et al.* (2010).

In most areas (SE, DG, IC, DJ, GG, GW, CB, CN, and JB), ozone concentration increasing is the highest in summer, and other areas (GJ, JN, GB, and JJ) show the highest ozone increasing in spring for 95th% ozone. For 67th% ozone, the highest ozone increasing trends are shown in summer with ratio (0.23-1.38) in IC, DJ, US, GG, GW, CB, CN, and JB. In all other areas except for BS, US and GN, the highest ozone increasing are shown in spring at 0.36-1.77. For 50th%, 33th%, 5th% ozone, the highest increasing ratio for each city and province in spring are absolutely higher (50th% 0.26-1.81; 33th% 0.28-1.59; 5th% 0.01-1.29) than those of other seasons (50th% 0.32-1.19; 33th% 0.97; 5th% −0.02-0.43).

3. 3 Investigation of the Main Factors Affecting Ozone Concentration

The factors affecting the trends of ozone concentration in each region were investigated using the results of the GLM. All factors (T_{max}, RH, WS_m, WS_a, T_{ds}, T_{d850}, T_{Dir}, and T_{Dis}) used in the GLM have impact on the ozone concentration. However, the main factors affecting the trends of ozone concentration are different in each region due to different characteristics of ozone formation or transport related with considering

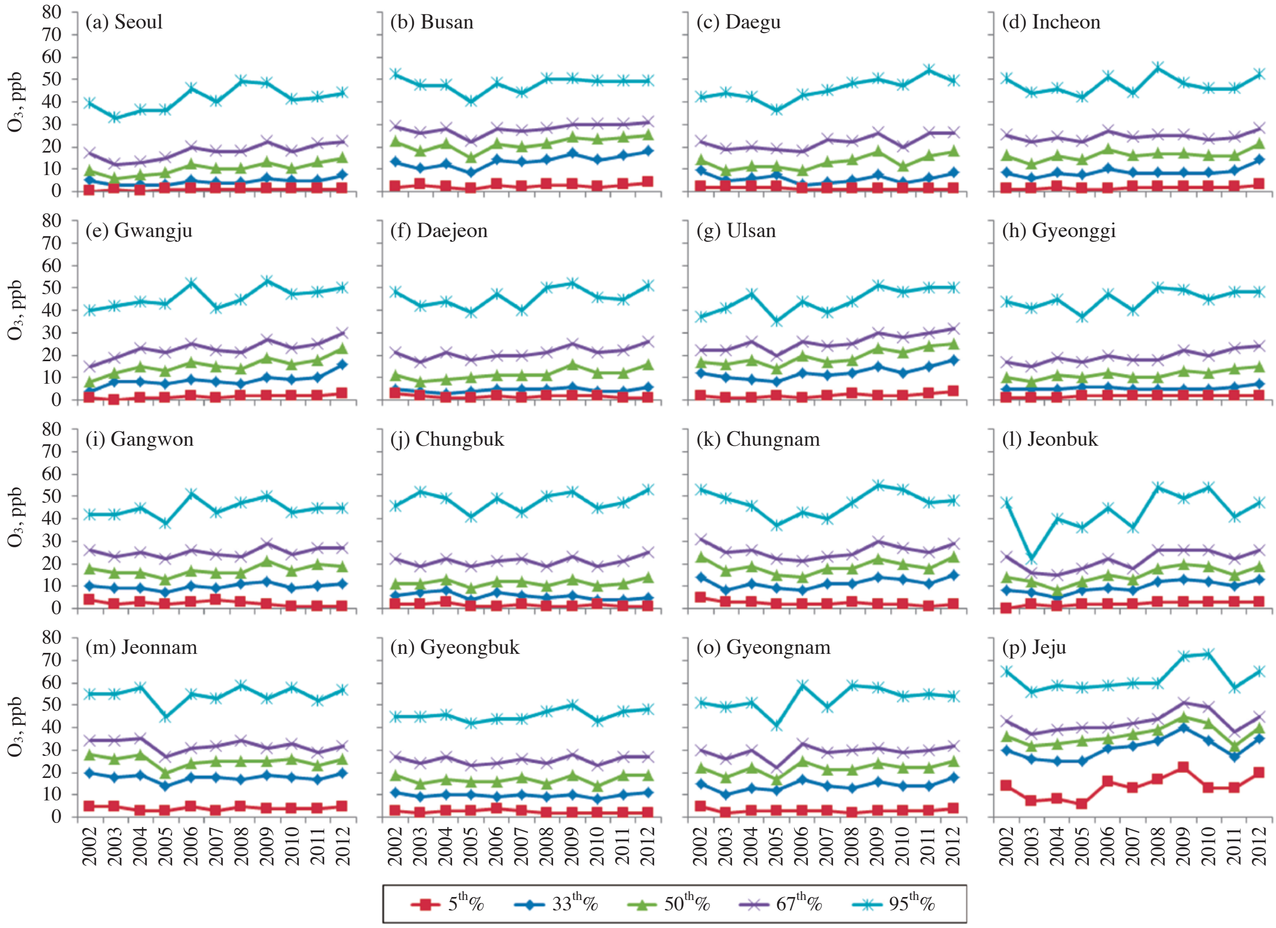

Fig. 6. Long term trends of percentile ozone of fall in each city and province (dark pink: 5th%; blue: 33th%, green: 50th%, purple: 67th%, sky blue: 95th%).

factors. This study chose top 2 factors, which is highly correlated with ozone concentration changes, determined by F-statistics in each region. Fig. 8 shows the main 2 factors affecting on the trends of ozone concentration in the 16 cities and province. The highest correlating factor with ozone in each city and province is shown in Fig. 8(a). Pink and yellow represent the region where the highest correlation factor is T_{max} and LRT related factors such as T_{Dir} and T_{Dis}, respectively. The second highest correlation factor with ozone in each city and province is shown in Fig. 8(b). Pink, yellow, green, and blue represent the region where the second highest correlating factor is T_{max}, transport related factors (T_{Dir} or T_{Dis}), RH, and wind speed (WS_m or WS_a), respectively.

In the case of the Seoul metropolitan area (SMA), which consists of Seoul, Incheon, and part of Gyeonggi province surrounding Seoul and Incheon, maximum temperature was the biggest factor affecting ozone concentration, and LRT related factors such as transport direction and transport distance was the biggest factor in other cities and province. The second biggest factor was relative humidity in Seoul and Incheon, LRT related factors in Gyeonggi, and relative humidity, maximum temperature, and wind speed in all other cities and province.

3.4 Extracting the Ozone Trends Adjusted Meteorologically and Precursor Emissions

This research calculated only the annual changes in ozone concentrations excluding meteorological fluctuations and long range transport fluctuations by stepwise applying the meteorological factors and LRT related factors by GLM. The meteorologically adjusted and LRT related factors adjusted long term trends of ozone in cities and province are shown in Fig. 9. Fig. 9 shows the annual changes in ozone concentrations (RAW) at the atmospheric monitoring stations in the 16 cities and province along with meteorologically and long

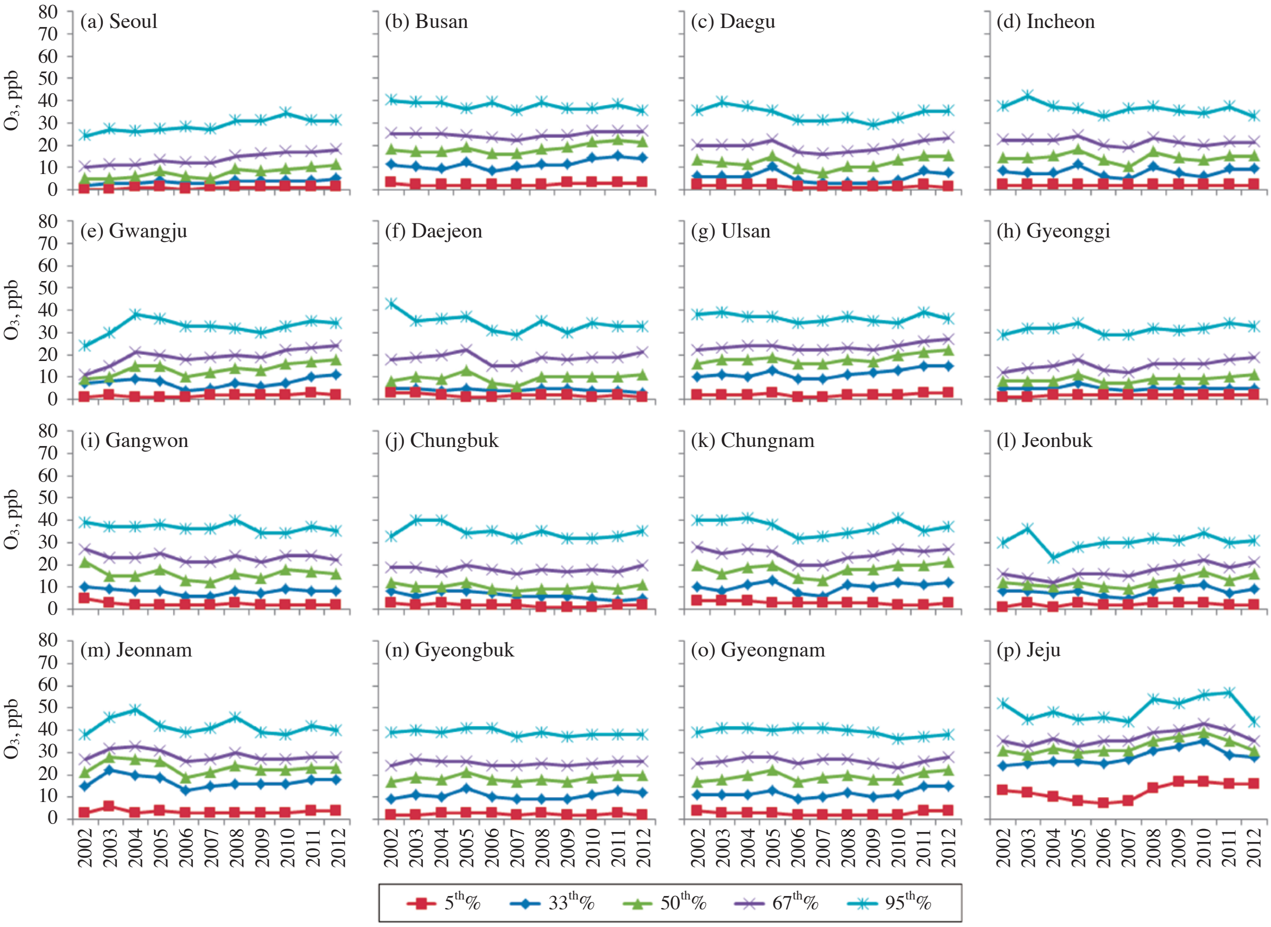

Fig. 7. Long term trends of percentile ozone of winter in each city and province (dark pink: 5th%; blue: 33th%, green: 50th%, purple: 67th%, sky blue: 95th%).

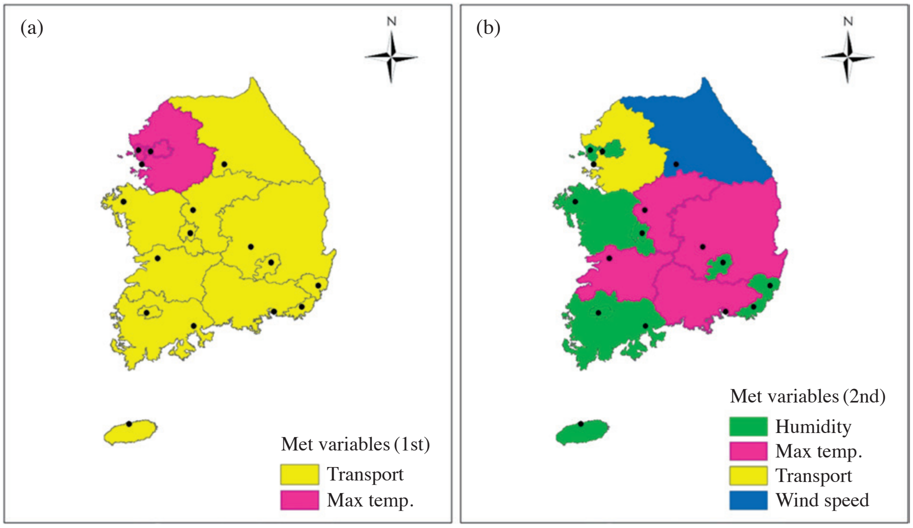

Fig. 8. The main influence factors affecting on the trends of ozone concentration; (a) the highest correlating factor, (b) the second highest correlating factor.

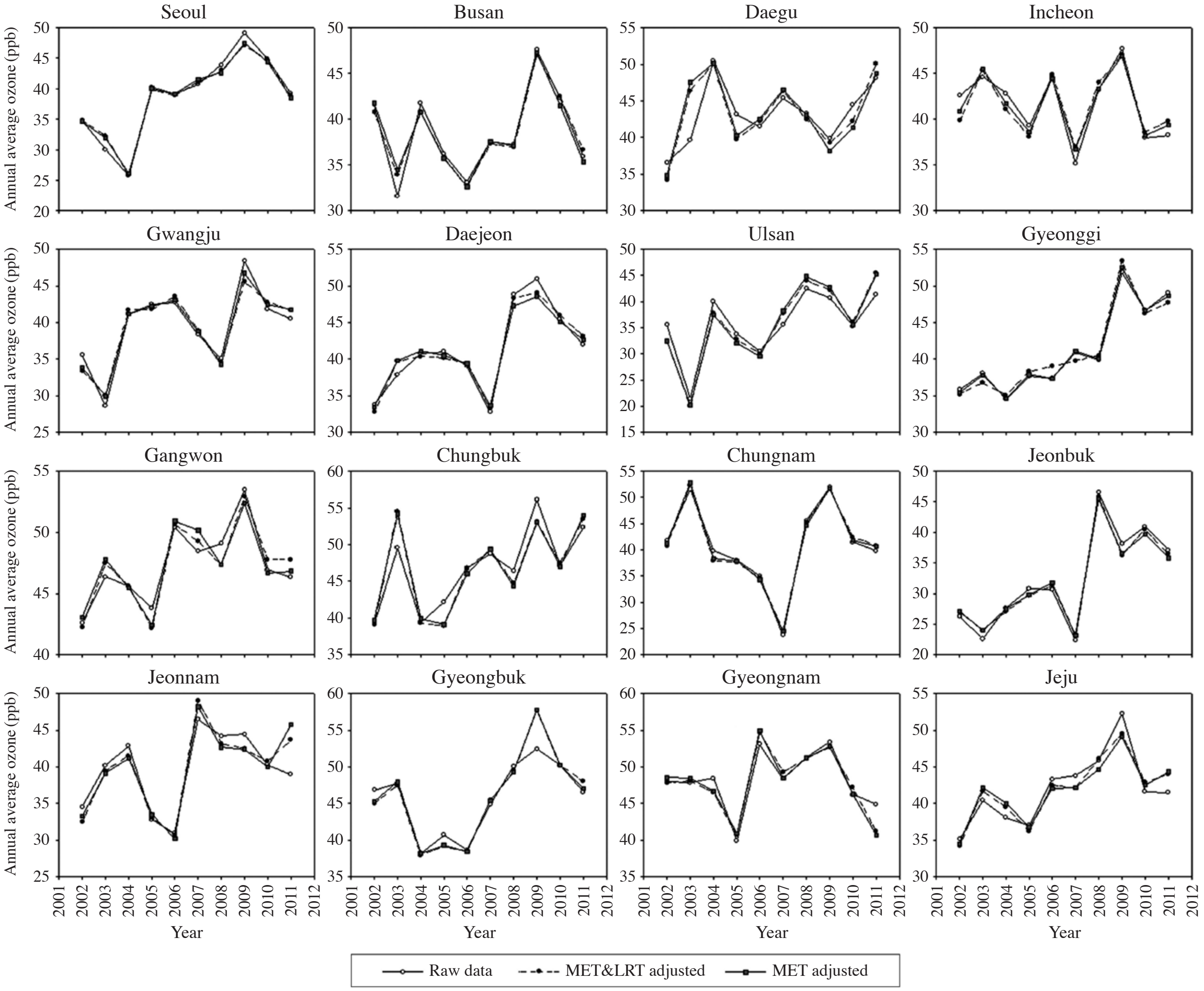

Fig. 9. The meteorologically adjusted and meteorologically & long range transport adjusted long term trends of ozone in each city and province.

range transport adjusted (MET&LRT adjusted) and only meteorologically adjusted (MET adjusted) datasets to investigate the effects of precursor emissions and LRT. The differences between ozone concentration (RAW) and adjusted concentrations were −7.6% and −6.2% as the largest difference in 2003 among 10 years for MET&LRT adjusted and MET adjusted concentration for Seoul, respectively. In the other cities and province, the differences between RAW and adjusted concentration (MET&LRT adjusted and MET adjusted) were relatively high in 2003. Year 2003 shows, excluding the effect of meteorological factors in almost regions, that ozone concentration increased in 11 city and province among all 16 areas, which leads to the conclusion that meteorological factors contributed to the decrease of ozone concentration. As such, anomaly detection of temperature and humidity, meteorological factors that have a high impact on ozone concentration, is shown in Fig. 10. Each anomaly is calculated as the difference between the average for the given year and the previous 9-year average. Negative value and positive value are lower and higher than average, respectively. Ambient temperature and relative humidity in year 2003 is lower and higher than those for previous 9-year, respectively. On the other hand, ambient temperature and relative humidity in year 2004 is higher than those for previous 9-year. Compared to year 2004, year 2003 showed considerably lower temperature (negative) and higher relative humidity (positive) nationwide. According to Camalier *et al.* (2007), 1% increase in relative humidity leads to a decrease in ozone by 0.5-1.5 ppb. When relative

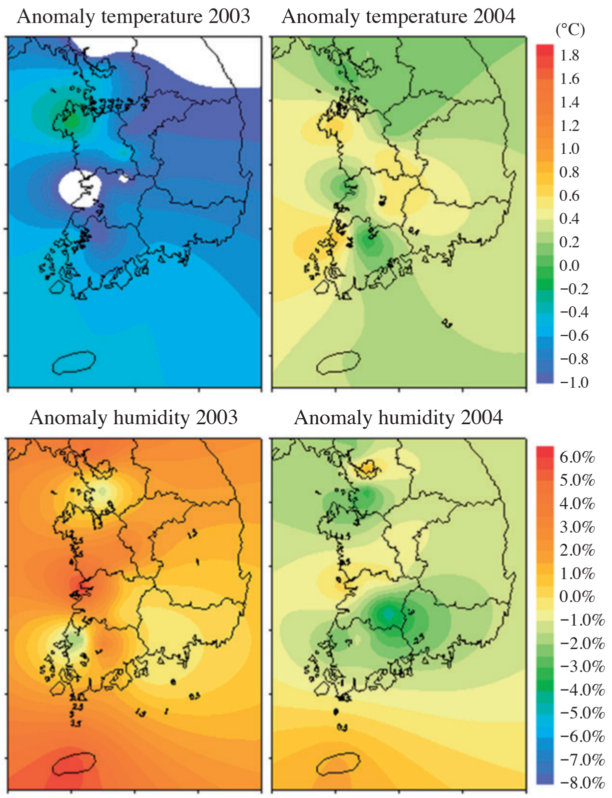

Fig. 10. Temperature and relative humidity anomalies for ozone seasons of 2003 and 2004.

humidity is high, the excited singlet (O(^{1}D)) oxygen atom, which is formed by ozone photolysis, produce more OH radical (Seinfeld and Pandis, 2006). The reactions of OH radical with NO_2 are higher than those with VOCs in VOCs-limited region such as Seoul (Shin *et al*., 2013). The OH-NO_2 reaction removes OH radicals from VOC oxidation cycle. Thus, the further ozone formation of ozone is retarded. Therefore, it can be inferred that the ozone formation was deterred by photochemical reaction due to low temperatures and high humidity, which has contributed to the decrease in ozone concentration.

4. SUMMARY AND CONCLUSION

In order to understand the current status of ozone pollution and establish effective public policies to reduce national ozone concentration, it is imperative to analyze the contribution of unintended factors affecting to the national ozone concentration. This study analyzed the overall trends of ozone concentrations in South Korea by performing statistical analysis of data collected from urban atmospheric monitoring stations throughout 2002-2011, and came to the following result and conclusion.

As observed in the results of the percentile concentration trends and KZ-filter analysis of this study, notwithstanding some differences in certain cities and province, the overall ozone levels in South Korea has been consistently increasing over the past 10 years. The ozone concentration in Seoul, the biggest city in Korea, is the lowest among concentration in other areas, but the increasing trends of ozone concentration is the highest in Korea for 95th% ozone. It is assumed that the ozone concentration in Seoul is the lowest because of NO_x titration due to the highest NO_x emissions in Korea. And the highest increasing ratio of higher ozone in Seoul could be coincidence with the fact that the ozone concentration in Seoul shows the highest correlation with maximum temperature by GLM analysis. It is thought that the active photochemical reaction could affect the higher ozone concentration increase.

On the other hands, the ozone concentration in Jeju is the highest in Korea with the highest increasing ratio for 5th%, 33th%, and 50th% ozone. It is also thought that the weak NO_x titration could be the reason of higher ozone concentration in Jeju. In case of Jeju, transport related factors is the major factor affecting the ozone trends. Thus, it is assumed that the variation of ozone trend of Asian region affecting the ozone trend in Jeju, where domestic ozone photochemical reaction is less active than urban area.

It is thought that the photochemical reaction plays the role of increasing ozone concentrations in the urban area, even though the LRT affected the increase of ozone concentration in non-urban area.

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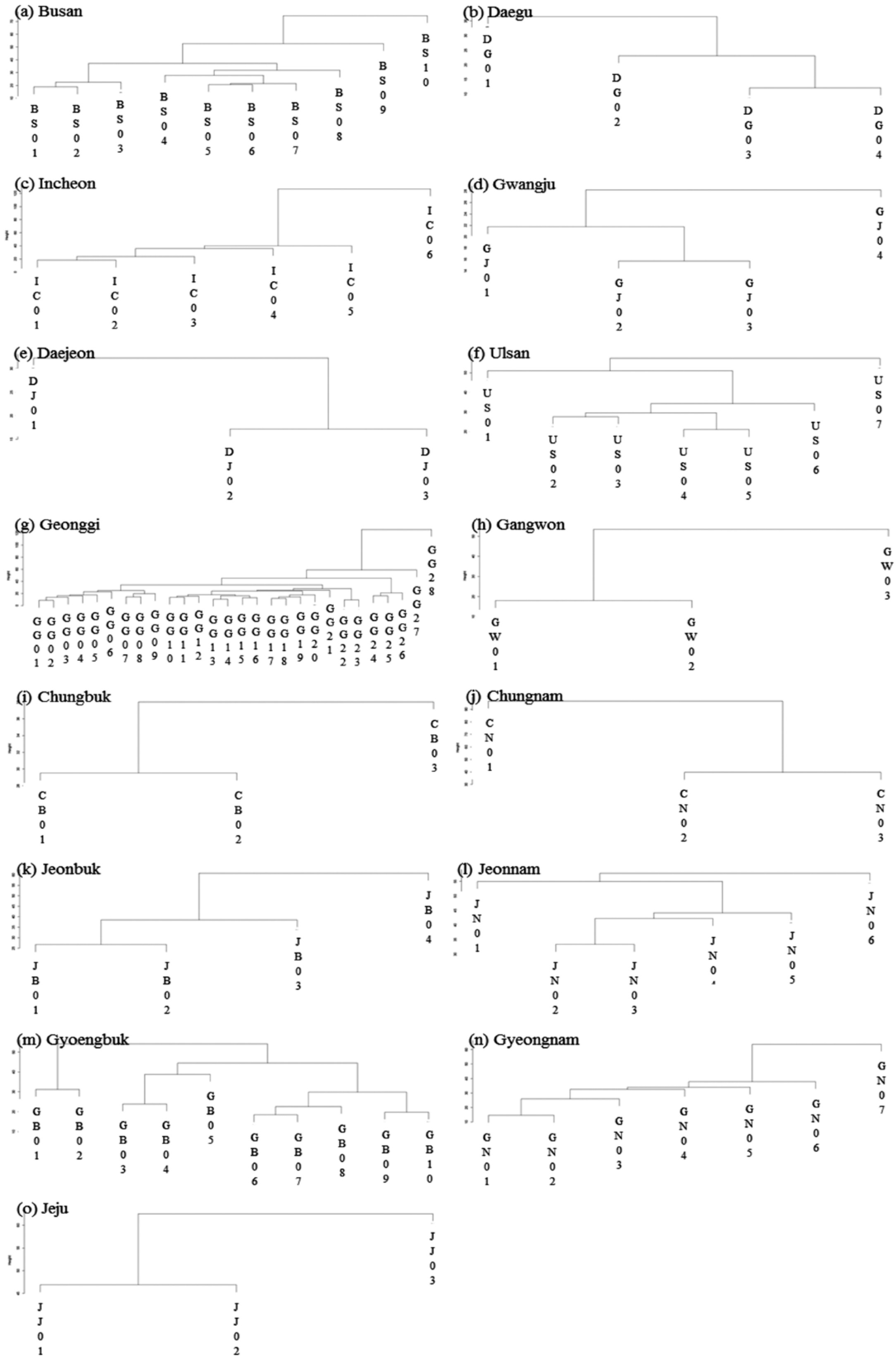
Table S1. The correlation coefficients of main affecting factors on the trends of ozone concentration.

	1st factor	2nd factor
Seoul (SE)	0.40	0.20
Busan (BS)	0.55	0.33
Daegu (DG)	0.61	0.54
Incheon (IC)	0.22	0.16
Gwangju (GJ)	0.40	0.20
Daejeon (DJ)	0.46	0.17
Ulsan (US)	0.37	0.11
Gyeonggi (GG)	0.27	0.13
Gangwon (GW)	0.35	0.31
Chungbuk (CB)	0.16	0.15
Chungnam (CN)	0.51	0.27
Jeonbuk (JB)	0.37	0.07
Jeonnam (JN)	0.34	0.31
Gyeongbuk (GB)	0.54	0.38
Gyeongnam (GN)	0.32	0.23
Jeju (JJ)	0.85	0.15



(a) Busan

(b) Daegu

(c) Incheon

(d) Gwangju

(e) Daejeon

(f) Ulsan

(g) Geonggi

(h) Gangwon

(i) Chungbuk

(j) Chungnam

(k) Jeonbuk

(l) Jeonnam

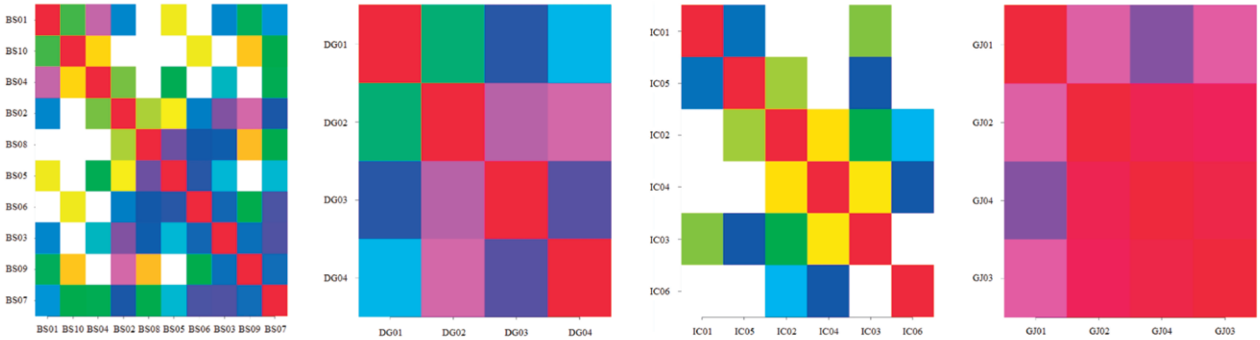
(m) Gyoengbuk

(n) Gyeongnam

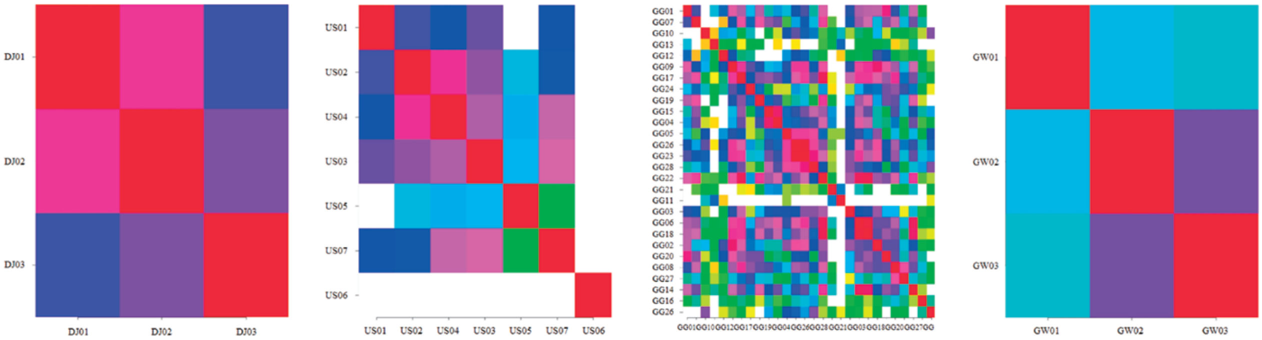
(o) Jeju

Fig. S1. The result of cluster analysis in other 15 cities and provinces for selecting representative monitoring stations.

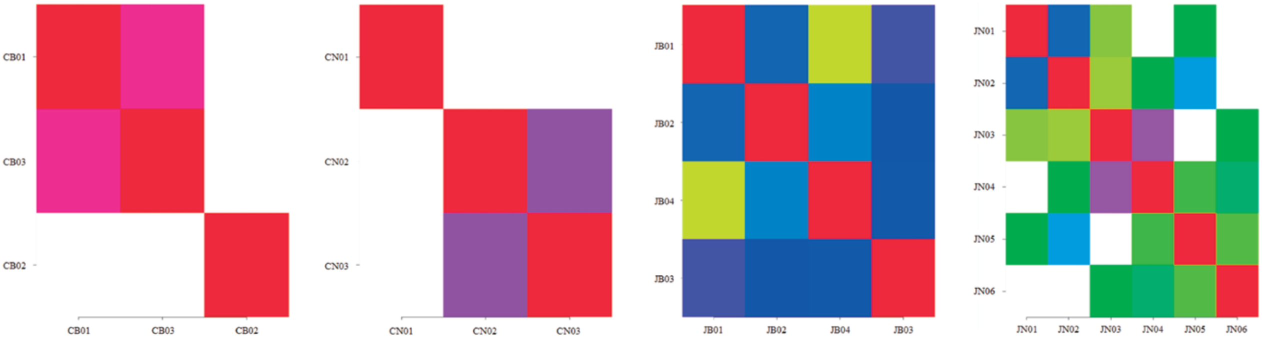
(a) Busan (b) Daegu (c) Incheon (d) Gwangju



(e) Daejeon (f) Ulsan (g) Gyeonggi (h) Gangwon



(i) Chungbuk (j) Chungnam (k) Jeonbuk (l) Jeonnam



(m) Gyeongbuk (n) Gyeongnam (o) Jeju

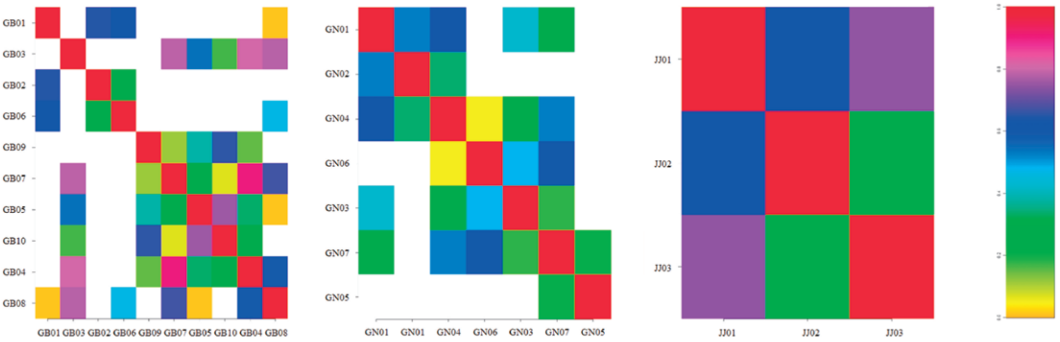


Fig. S2. The result of correlation matrix in other 15 cities and provinces for selecting representative monitoring stations.