

Table 1. List of acronyms used in this study.

Acronyms	Original words	Details
R	Residence	Residential Areas : Areas necessary to protect peaceful dwelling and sound living environment
C	Commerce	Commercial Areas : Areas necessary to increase convenience in commerce and other businesses
I	Industry	Industrial Areas : Areas necessary to increase convenience of industries
G	Greenbelt	Green Areas : Areas requiring the conservation of green areas to protect the natural environment, farmland and forests, health and sanitation, security and to prevent any disorderly sprawl of cities
SMA	Seoul Metropolitan Area	
CNSP	CO, NO <sub>2</sub> , SO <sub>2</sub> and PM <sub>10</sub>	
OZIPR	Ozone Isopleth Plotting Package for Research	
MEK	Ministry of Environment of Korea	
MLIT	Ministry of Land, Infrastructure and Transport	
AVHRR	Advanced Very High Resolution Radiometer	
MODIS	Moderate-resolution Imaging Spectroradiometer	

Table 2. Data information of the surface air pollutants (O<sub>3</sub>, CO, NO<sub>2</sub>, SO<sub>2</sub> and PM<sub>10</sub>) measured at 283 air pollution monitoring stations of the Ministry of Environment of Korea (MEK) in South Korea during 2002-2013. The information for the VOCs at 9 of the photochemical MEK stations, simultaneously measured with the other pollutants at the same sites, has also been shown. The 9 out of the total 19 VOCs stations were selected in this study, based on their locations and their relatively long-term records since 2007.

Air pollutant	Source	Period	Time interval	Number of stations				
				Residence	Commerce	Industry	Greenbelt	Total
O <sub>3</sub> , CO, NO <sub>2</sub> , SO <sub>2</sub> , PM <sub>10</sub>	MEK	Jan 2002 – Dec 2013	Hourly	154	57	35	37	283
VOCs	MEK	Jan 2007 – Dec 2013	Hourly	3	1	0	3	7
VOCs at Daemyoung (128.57E, 35.84N)	MEK	Jan 2010 – Dec 2013	Hourly	1	0	0	0	1
VOCs at Joongheung (126.68E, 34.83N)	MEK	Jan 2008 – Dec 2013	Hourly	0	0	1	0	1

Table 3. Methods and instruments for measuring the surface air pollutants ( $O_3$ , CO,  $NO_2$ ,  $SO_2$  and  $PM_{10}$ ) at 283 MEK air pollution monitoring stations in South Korea during 2002-2013.

Air Pollutant	Method	Instrument
$O_3$	U.V Photometric Method	Thermo, 49i
CO	Non-Dispersive Infrared Method	Thermo, 48CTL
$NO_2$	Chemiluminescent Method	Thermo, 42CTL
$SO_2$	Pulse U.V Fluorescence Method	Thermo, 43CTL
$PM_{10}$	$\beta$ -ray Absorption Method	Thermo, FH62-C14
VOC	TD-GC/MS (Thermal Desorption Gas Chromatography/Mass Spectrometry)	Agilent, Perkinelmer, Varian

Table 4. Comparison of the four land-use types of the MEK (residence, commerce, industry, and greenbelt) for 283 air pollution monitoring stations of the MEK during 2002-2012 with the satellite-derived land-cover types of the AVHRR and MODIS in a  $0.25^\circ \times 0.25^\circ$  grid. The AVHRR data were available for 13 land-cover types over the globe at a 1km x 1km pixel resolution during 1981-1994 (e.g., De Fries et al., 1998; Hansen et al., 2000). The MODIS data have been derived for 17 land-cover types over the globe at a 5km x 5km spatial resolution during 2002-2012 (e.g., Friedl et al., 2010). In this study, for comparison, the AVHRR and MODIS original types were regrouped into the following four land-cover types: forest/wood, grass/shrub, urban/built-up and water. In the table, the values with and without parentheses indicate the MODIS and AVHRR data, respectively.

Land Cover	Residence (%)	Commerce (%)	Industry (%)	Greenbelt (%)
Forest/Wood	12.4 (31.8)	15.8 (35.8)	8.6 (26.2)	35.2 (37.2)
Grass/Shrub	58.4 (27.5)	43.9 (18.0)	60.0 (19.2)	43.2 (21.8)
Urban/Built-up	19.5 (28.8)	33.3 (32.2)	11.4 (28.6)	0.0 (16.4)
Water	9.7 (11.9)	7.0 (14.0)	20.0 (26.0)	21.6 (24.6)

Table 5. Climatological averages of (a)  $O_3$  (ppb), (b) CO (0.1 ppm), (c)  $NO_2$  (ppb), (d)  $SO_2$  (ppb), and (e)  $PM_{10}$  ( $\mu g m^{-3}$ ) in two types of spatial grids ( $0.25^\circ \times 0.25^\circ$  and  $0.1^\circ \times 0.1^\circ$ ) over South Korea during 2002-2013. The standard deviation ( $\sigma$ ) values of the five kinds of variables are also presented with the  $\pm$  values.

	Spring	Summer	Fall	Winter	Annual
<b>&lt; <math>0.25^\circ \times 0.25^\circ</math> &gt;</b>					
$O_3$ (ppb)	34.93 $\pm$ 7.69	27.22 $\pm$ 4.44	22.34 $\pm$ 7.22	19.62 $\pm$ 7.08	26.08 $\pm$ 6.33
CO (0.1ppm)	5.38 $\pm$ 1.12	4.16 $\pm$ 0.92	5.41 $\pm$ 1.24	7.23 $\pm$ 2.05	5.53 $\pm$ 1.23
$NO_2$ (ppb)	18.10 $\pm$ 8.25	13.14 $\pm$ 6.25	17.92 $\pm$ 8.35	21.13 $\pm$ 9.01	17.54 $\pm$ 7.88
$SO_2$ (ppb)	4.89 $\pm$ 1.65	3.57 $\pm$ 1.71	4.23 $\pm$ 1.61	6.49 $\pm$ 2.43	4.78 $\pm$ 1.67
$PM_{10}$ ( $\mu g m^{-3}$ )	64.47 $\pm$ 8.41	41.2 $\pm$ 6.21	45.57 $\pm$ 7.49	54.82 $\pm$ 10.89	51.46 $\pm$ 7.72
<b>&lt; <math>0.1^\circ \times 0.1^\circ</math> &gt;</b>					
$O_3$ (ppb)	33.08 $\pm$ 7.37	26.42 $\pm$ 4.24	20.87 $\pm$ 6.51	17.95 $\pm$ 6.59	24.63 $\pm$ 5.88
CO (0.1ppm)	5.38 $\pm$ 1.14	4.21 $\pm$ 0.97	5.49 $\pm$ 1.30	7.30 $\pm$ 2.06	5.58 $\pm$ 1.27
$NO_2$ (ppb)	21.10 $\pm$ 9.59	15.48 $\pm$ 7.42	20.79 $\pm$ 9.27	24.12 $\pm$ 9.99	20.34 $\pm$ 8.96
$SO_2$ (ppb)	5.27 $\pm$ 1.97	3.97 $\pm$ 2.10	4.60 $\pm$ 1.86	6.82 $\pm$ 2.45	5.15 $\pm$ 1.90
$PM_{10}$ ( $\mu g m^{-3}$ )	66.53 $\pm$ 9.90	42.91 $\pm$ 7.01	47.63 $\pm$ 8.53	57.31 $\pm$ 12.30	53.58 $\pm$ 8.91

Table 6. The spatial mean and standard deviation of the surface air pollutant concentration averages ( $O_3$ , CO,  $NO_2$ ,  $SO_2$ , and  $PM_{10}$ ) in the diurnal, weekly, and annual variations over South Korea during 2002-2013 in a  $0.25^\circ \times 0.25^\circ$  grid in terms of the four land-use types of MEK as follows: residence (R), commerce (C), industry (I), and greenbelt (G). Here the values in parentheses denote the mean and standard deviation in a  $0.1^\circ \times 0.1^\circ$  grid.

Cycle and pollutants	Residence		Commerce		Industry		Greenbelt	
<b>Diurnal</b>								
O <sub>3</sub>	24.3±8.07	(23.5±8.19)	21.3±6.93	(20.2±6.80)	23.5±7.24	(23.5±7.20)	30.9±7.69	(30.4±7.78)
CO	5.7±0.56	(5.7±0.60)	6.2±0.63	(6.4±0.60)	5.7±0.39	(5.8±0.42)	4.6±0.26	(4.7±0.28)
NO <sub>2</sub>	21.1±3.62	(23.1±3.87)	25.1±4.19	(28.1±4.33)	23.2±3.02	(23.8±2.98)	11.7±1.52	(12.7±1.70)
SO <sub>2</sub>	5.2±0.33	(5.3±0.35)	5.6±0.39	(5.7±0.41)	6.8±0.79	(7.5±0.85)	3.3±0.23	(3.4±0.24)
PM <sub>10</sub>	52.7±3.04	(53.3±2.87)	54.0±3.37	(55.2±3.28)	56.0±2.98	(56.4±3.01)	48.4±2.20	(49.5±2.33)
<b>Weekly</b>								
O <sub>3</sub>	24.2±0.72	(23.4±0.81)	21.2±0.75	(20.2±0.84)	23.4±1.19	(23.4±1.22)	30.8±0.41	(30.3±0.46)
CO	5.7±0.01	(5.7±0.11)	6.2±0.16	(6.4±0.18)	5.7±0.14	(5.8±0.14)	4.6±0.01	(4.7±0.01)
NO <sub>2</sub>	21.1±1.32	(23.1±1.48)	25.1±1.42	(28.2±1.65)	23.2±1.99	(23.8±2.03)	11.7±0.69	(12.7±0.78)
SO <sub>2</sub>	5.2±0.15	(5.3±0.15)	5.5±0.12	(5.7±0.15)	6.8±0.29	(7.5±0.30)	3.3±0.02	(3.4±0.01)
PM <sub>10</sub>	52.7±1.19	(53.3±1.31)	54.0±1.20	(55.2±1.43)	56.1±2.25	(56.4±2.25)	48.4±0.71	(49.4±0.82)
<b>Annual</b>								
O <sub>3</sub>	24.0±6.96	(23.4±6.89)	20.9±6.25	(20.2±6.12)	23.2±6.23	(23.4±6.28)	30.7±7.35	(30.3±7.34)
CO	5.8±1.32	(5.7±1.30)	6.3±1.42	(6.4±1.36)	5.8±0.93	(5.8±0.93)	4.7±0.89	(4.7±0.92)
NO <sub>2</sub>	21.1±4.01	(23.2±4.27)	25.2±3.79	(28.2±3.91)	23.2±3.47	(23.8±3.55)	11.7±2.37	(12.8±2.59)
SO <sub>2</sub>	5.2±1.28	(5.3±1.23)	5.6±1.39	(5.7±1.31)	6.8±0.87	(7.5±0.76)	3.4±0.92	(3.4±0.93)
PM <sub>10</sub>	53.1±10.40	(53.3±10.45)	54.5±10.93	(55.2±10.83)	56.4±9.68	(56.3±9.45)	48.8±9.85	(49.6±9.82)

Table 7. The magnitude order of the surface air pollutant concentration averages ( $O_3$ , CO,  $NO_2$ ,  $SO_2$ , and  $PM_{10}$ ) in the diurnal, weekly and annual variations of Fig. 6 over South Korea during 2002-2013 in a  $0.25^\circ \times 0.25^\circ$  grid in terms of the four land-use types of MEK as follows: residence (R), commerce (C), industry (I) and greenbelt (G). The numbers in the table indicate the ranking of each pollutant, based on the pollutant concentration values over the types. Here the greater concentration, the higher ranking. If the orders in the two grids are different from each other, then those in parentheses have been shown for the  $0.1^\circ \times 0.1^\circ$  grid.

Cycle/pollutants	Residence	Commerce	Industry	Greenbelt	Order
<b>Diurnal</b>					
$O_3$	2 (2)	4 (4)	3 (3)	1 (1)	G>R>I>C
CO	2 (3)	1 (1)	3 (2)	4 (4)	C>R>I>G (C>I>R>G)
$NO_2$	3 (3)	1 (1)	2 (2)	4 (4)	C>I>R>G
$SO_2$	3 (3)	2 (2)	1 (1)	4 (4)	I>C>R>G
$PM_{10}$	3 (3)	2 (2)	1 (1)	4 (4)	I>C>R>G
<b>Weekly</b>					
$O_3$	2 (2)	4 (4)	3 (3)	1 (1)	G>R>I>C
CO	2 (3)	1 (1)	3 (2)	4 (4)	C>R>I>G (C>I>R>G)
$NO_2$	3 (3)	1 (1)	2 (2)	4 (4)	C>I>R>G
$SO_2$	3 (3)	2 (2)	1 (1)	4 (4)	I>C>R>G
$PM_{10}$	3 (3)	2 (2)	1 (1)	4 (4)	I>C>R>G
<b>Annual</b>					
$O_3$	2 (2)	4 (4)	3 (3)	1 (1)	G>R>I>C
CO	2 (3)	1 (1)	3 (2)	4 (4)	C>R>I>G (C>I>R>G)
$NO_2$	3 (3)	1 (1)	2 (2)	4 (4)	C>I>R>G
$SO_2$	3 (3)	2 (2)	1 (1)	4 (4)	I>C>R>G
$PM_{10}$	3 (3)	2 (2)	1 (1)	4 (4)	I>C>R>G

121 Table 8. Comparisons of the climatological annual averages over South Korea during 2002-2013, based on the two types of spatial  
 122 scale analyses of the 0.1°×0.1° and 0.25°×0.25° grids. The 0.1°×0.1° grid averages (compared to those of 0.25°×0.25°) generally  
 123 tend to show the characteristics in big urban cities rather than in suburban small suburban cities, because the air-pollution  
 124 monitoring stations are more densely located in the former areas.

Air pollutant	Average (0.1°×0.1°) minus Average (0.25°×0.25°)			
	Residence	Commerce	Industry	Greenbelt
O <sub>3</sub> (ppb)	-0.513	-0.735	0.181	-0.342
CO (0.1 ppm)	-0.067	0.093	0.052	0.009
NO <sub>2</sub> (ppb)	2.020	2.969	0.573	0.767
SO <sub>2</sub> (ppb)	0.036	0.123	0.687	0.033
PM <sub>10</sub> (μg m <sup>-3</sup> )	0.270	0.711	-0.012	0.409

Table 9. Trends of surface air pollutants (O<sub>3</sub>, NO<sub>2</sub>, OX, CO, SO<sub>2</sub> and PM<sub>10</sub>) over South Korea during 2002-2013, based on the three types of analyses (0.1°×0.1° grid and 0.25°×0.25° grid) over the four land-use types of the MEK of residence (R), commerce (C), industry (I), and greenbelt (G). The magnitude order for the trends of each of the pollutant over the types has been shown in the figures. The ± trend values indicate the 95% confidence intervals. It should be noted that the trend values are statistically significant except for some of the NO<sub>2</sub> and SO<sub>2</sub> cases, marked by an asterisk(\*).

	Residence	Commerce	Industry	Greenbelt	Trend	Order
<b>0.25°×0.25°</b>						
O <sub>3</sub> (ppb yr <sup>-1</sup> )	0.501±0.098	0.407±0.095	0.352±0.093	0.369±0.094	increase	R>C>G>I
NO <sub>2</sub> (ppb yr <sup>-1</sup> )	-0.295±0.081	-0.042±0.088*	-0.135±0.084	-0.100±0.053	decrease	R>I>G>C*
OX (ppb yr <sup>-1</sup> )	0.205±0.107	0.365±0.103	0.231±0.113	0.260±0.103	increase	C>G>I>R
CO (0.1ppm yr <sup>-1</sup> )	-0.202±0.021	-0.210±0.021	-0.247±0.025	-0.135±0.022	decrease	I>C>R>G
SO <sub>2</sub> (ppb yr <sup>-1</sup> )	-0.036±0.024	-0.114±0.028	-0.140±0.029	-0.060±0.016	decrease	I>C>G>R
PM <sub>10</sub> (μgm <sup>-3</sup> yr <sup>-1</sup> )	-1.038±0.459	-1.014±0.456	-1.003±0.480	-1.098±0.485	decrease	G>R>C>I
<b>0.1°×0.1°</b>						
O <sub>3</sub> (ppb yr <sup>-1</sup> )	0.545±0.096	0.462±0.092	0.340±0.094	0.326±0.095	increase	R>C>I>G
NO <sub>2</sub> (ppb yr <sup>-1</sup> )	-0.240±0.083	-0.078±0.092*	-0.054±0.084*	-0.023±0.054*	decrease	R>C*>I*>G*
OX (ppb yr <sup>-1</sup> )	0.304±0.108	0.396±0.106	0.299±0.110	0.300±0.102	increase	C>R>G>I
CO (0.1ppm yr <sup>-1</sup> )	-0.175±0.021	-0.204±0.020	-0.246±0.025	-0.124±0.022	decrease	I>C>R>G
SO <sub>2</sub> (ppb yr <sup>-1</sup> )	-0.019±0.023*	-0.104±0.027	-0.177±0.030	-0.050±0.015	decrease	I>C>G>R
PM <sub>10</sub> (μgm <sup>-3</sup> yr <sup>-1</sup> )	-1.374±0.535	-1.290±0.474	-0.926±0.492	-1.049±0.485	decrease	R>C>G>I

Table 10. Climatological average value of O<sub>3</sub> (ppb) and NO<sub>2</sub> (ppb) over South Korea during 2002-2013 in terms of the MEK four land-use types (residence, commerce, industry and greenbelt) over the 283 total stations and the 209 stations excluding the 74 SMA residence areas, respectively. The spatial variation in the pollutant concentrations for the individual type is presented with the standard deviation of ± values. The number in the parenthesis indicates the ranking of each pollutant, based on concentration value over the type.

		All stations		Stations excluding the SMA residence areas	
Land-use type	Air Pollutant	O <sub>3</sub>	NO <sub>2</sub>	O <sub>3</sub>	NO <sub>2</sub>
<b>&lt;Annual&gt;</b>					
Residence		22.4±4.32 (3)	25.9±8.15 (2)	25.0±4.03 (2)	20.3±4.94 (3)
Commerce		19.2±4.85 (4)	31.3±12.00 (1)		
Industry		23.4±4.32 (2)	24.3±6.89 (3)	23.4±4.32 (3)	24.3±6.89 (2)
Greenbelt		30.2±7.83 (1)	13.3±9.63 (4)		
<b>&lt;Winter only&gt;</b>					
Residence		15.4±4.91 (3)	30.6±9.04 (2)	18.3±4.80 (2)	24.4±6.00 (3)
Commerce		13.2±4.28 (4)	34.6±11.29(1)		
Industry		16.7±4.43 (2)	27.8±7.52 (3)	16.7±4.43 (3)	27.8±7.52 (2)
Greenbelt		24.4±8.72 (1)	16.3±11.27(4)		

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Table 11. The spatial mean values of the long-term surface air pollutant concentration averages (O<sub>3</sub>, NO<sub>2</sub>, OX, VOC, and VOC/NO<sub>2</sub>) at 9 of the photochemical air pollution monitoring stations of the MEK over South Korea since 2007 in terms of the four MEK land-use categories as follows: residence (R), commerce (C), industry (I), and greenbelt (G).

Air pollutant	Residence	Commerce	Industry	Greenbelt	Average
O <sub>3</sub> (ppb)	21.8±1.24	22.0	31.0	35.5±0.43	27.6
NO <sub>2</sub> (ppb)	31.8±2.18	35.5	19.7	9.9±0.39	24.2
OX=O <sub>3</sub> +NO <sub>2</sub> (ppb)	53.6±1.00	57.4	50.7	45.4±0.29	51.8
VOC (ppbC)	112.2±14.84	308.3	199.6	31.2±2.14	162.8
VOC/NO <sub>2</sub>	3.6±0.27	8.7	10.2	3.9±0.13	6.6