

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 ₂ , SO ₂ and PM ₁₀ | |
| 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 (O₃, CO, NO₂, SO₂ and PM₁₀) 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 ₃ , CO, NO ₂ , SO ₂ , PM ₁₀ | 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 (O₃, CO, NO₂, SO₂ and PM₁₀) 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 | β-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° x 0.25° 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) O₃ (ppb), (b) CO (0.1 ppm), (c) NO₂ (ppb), (d) SO₂ (ppb), and (e) PM₁₀ (μgm⁻³) in two types of spatial grids (0.25°x0.25° and 0.1°x0.1°) over South Korea during 2002-2013. The standard deviation (σ) values of the five kinds of variables are also presented with the ± values.

| | Spring | Summer | Fall | Winter | Annual |
|---------------------------------------|------------|------------|------------|-------------|------------|
| < 0.25°x0.25°> | | | | | |
| O ₃ (ppb) | 34.93±7.69 | 27.22±4.44 | 22.34±7.22 | 19.62±7.08 | 26.08±6.33 |
| CO (0.1ppm) | 5.38±1.12 | 4.16±0.92 | 5.41±1.24 | 7.23±2.05 | 5.53±1.23 |
| NO ₂ (ppb) | 18.10±8.25 | 13.14±6.25 | 17.92±8.35 | 21.13±9.01 | 17.54±7.88 |
| SO ₂ (ppb) | 4.89±1.65 | 3.57±1.71 | 4.23±1.61 | 6.49±2.43 | 4.78±1.67 |
| PM ₁₀ (μgm ⁻³) | 64.47±8.41 | 41.2±6.21 | 45.57±7.49 | 54.82±10.89 | 51.46±7.72 |
| < 0.1°x0.1°> | | | | | |
| O ₃ (ppb) | 33.08±7.37 | 26.42±4.24 | 20.87±6.51 | 17.95±6.59 | 24.63±5.88 |
| CO (0.1ppm) | 5.38±1.14 | 4.21±0.97 | 5.49±1.30 | 7.30±2.06 | 5.58±1.27 |
| NO ₂ (ppb) | 21.10±9.59 | 15.48±7.42 | 20.79±9.27 | 24.12±9.99 | 20.34±8.96 |
| SO ₂ (ppb) | 5.27±1.97 | 3.97±2.10 | 4.60±1.86 | 6.82±2.45 | 5.15±1.90 |
| PM ₁₀ (μgm ⁻³) | 66.53±9.90 | 42.91±7.01 | 47.63±8.53 | 57.31±12.30 | 53.58±8.91 |

메모 [YJH1]: Referee#2, A7
(removing yr-1)

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.

메모 [YJH2]: Referee#3, A11

| Cycle and pollutants | Residence | | Commerce | | Industry | | Greenbelt | |
|----------------------|------------|--------------|------------|--------------|-----------|-------------|-----------|-------------|
| Diurnal | | | | | | | | |
| O ₃ | 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 ₂ | 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 ₂ | 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 ₁₀ | 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) |
| Weekly | | | | | | | | |
| O ₃ | 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 ₂ | 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 ₂ | 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 ₁₀ | 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) |
| Annual | | | | | | | | |
| O ₃ | 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 ₂ | 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 ₂ | 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 ₁₀ | 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.

메모 [YJH3]: Referee#3, A11

| Cycle/pollutants | Residence | Commerce | Industry | Greenbelt | Order |
|------------------|-----------|----------|----------|-----------|-------------------|
| Diurnal | | | | | |
| 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 |
| Weekly | | | | | |
| 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 |
| Annual | | | | | |
| 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 |

Table 8. Comparisons of the climatological annual averages over South Korea during 2002-2013, based on the two types of spatial 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 tend to show the characteristics in big urban cities rather than in suburban small suburban cities, because the air-pollution 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 ₃ (ppb) | -0.513 | -0.735 | 0.181 | -0.342 |
| CO (0.1 ppm) | -0.067 | 0.093 | 0.052 | 0.009 |
| NO ₂ (ppb) | 2.020 | 2.969 | 0.573 | 0.767 |
| SO ₂ (ppb) | 0.036 | 0.123 | 0.687 | 0.033 |
| PM ₁₀ (μg m ⁻³) | 0.270 | 0.711 | -0.012 | 0.409 |

Table 11. The spatial mean values of the long-term surface air pollutant concentration averages (O_3 , NO_2 , OX , VOC , and VOC/NO_2) 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).

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